Natural Dye Extracted from *Dalbergia cochinchinensis* Residue with Water Fastness, Mildew Resistance and Permeability Properties for Wood Staining

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Research Article

Keywords: Dalbergia cochinchinensis extracts, Natural dye, Water fastness, Mildew resistance, Permeability

Posted Date: August 27th, 2021

DOI: https://doi.org/10.21203/rs.3.rs-848625/v1

// Abstract

Wood dyed with extracts from *Dalbergia cohinchinensis* residues as natural dye (ND) exhibited desirable color appearance and anti-UV property while the potential property of the ND dyed wood has not been exploited with respect to the water fastness, mildew resistance along with the penetrability of the ND on wood blocks dyeing. The present study was aimed at exploring these aspects for multifunction assessment of the ND dyed wood. The results showed that the ΔE^* (4.58) and color intensity reduction (PR:14.01%) of the ND dyed wood declined slightly after washed treatment by comparison with that of acid red (ΔE^*:32.82; RP:76.14%) and reactive red (ΔE^*:26.85; RP:66.52%) dyed wood indicative of its preferable waster fastness due to the surface hydrophobicity improvement. In addition, the ND ameliorated the mildew resistance against *Aspergillus niger* and *Trichoderma viride* infection. Interestingly, the wood blocks inner can be completely impregnated with ND under atmospheric pressure dyeing process without any pretreatment and auxiliary adding. The extracts utilization scenario was proved to perforce greatly promising for industrial application in terms of multifunctional ND dyed wood preparation.

1. Introduction

ND exploitation garners increasing attention ascribed to its expected property regarding eco-friendly, non-hazardous, biodegradability and compatible with environment, etc (Haji and Naebe, 2020). The history of hunt for ND usage was more than 4000 years since ancient time (Canevari et al., 2016). The available ND was mainly extracted from various substances including plants, insects, shellfish and even from lichens at that period (Rosenberg, 2008). Up to date, researchers have principally focused their interests in usage of extracts from different plants as ND due to plenty of chromophore in the extracts, which can endow the dyed materials with different color shades by reflecting and absorbing the visible lights (Falkehag. et al., 1966; K. et al., 2020; Zhou et al., 2020). Interestingly, several ND can impart the dyed textile with multi-functional property pertain to appealing anti-UV and antibacterial performance, which was probably owing to ample natural phenols and/or flavonoid compounds existed in the extracts (Bhuiyan et al., 2017; Rather et al., 2020; Silva et al., 2018). Additionally, literatures have reported that extracts from heartwood can be used as natural photo-stabilizers and wood preservative against termite (Brocco et al., 2019; Chang et al., 2015).

Generally, wood extracts exhibit finely affinity with its color appearance and some dark-colored wood invariably have higher content of phenolic components (Dellus et al., 1997; Dünisch et al., 2009). Accordingly, its biological resistance is ameliorated pronouncedly compared with sapwood and it is undoubtedly favored by more consumers in wooden products market. One crucial fact should be realized that longer growth cycle of this kind of wood is needed, which can't meet extensive demands of wooden products in peoples' daily life. Thereby fast-grown wood (*poplar* spp.) have been chosen as potential candidate for furniture production and indoor decoration. It is notable that one of the most drawbacks directly from peoples' perception *i.e.* poor color appearance of this kind wood limited its utilization. Up to date, dyeing process (mainly organic synthetic dye) is served as effective means for solving this problem in a greater degree due to it can endow fast-grown wood substrate with manifold color shades (Liu et al., 2015b; Liu et al., 2015c). Nevertheless, this kind of dyed wood are always prone to terrible discoloration when subject to solar irradiation (Liu et al., 2015a). As for this issue, in our pervious study, a sustainable and eco-friendly dyeing method has been exploited and the issues aforementioned have been solved successfully by employing *D. cochinchinensis* extracts as ND to impart the *poplar* veneers with evenly color appearance (yellowish-brown) and appreciable anti-UV property (Zhu et al., 2021). Unquestionably, this treatment strategy can improve the value-added of *poplar* wood by overcoming its some inherent demerits. However, the water fastness and mildew resistance property of this ND dyed wood are still unknown and further studies are needed for these aspects. In addition, wood blocks dyeing in-depth under atmospheric pressure is extremely tough due to its pronounced hierarchical anisotropy at multiple length scales (Chen et al., 2020; Ling et al., 2018; Zhu et al., 2016). An attempt exploration was also conducted on this issue by employed ND to dye wood blocks in anhydrous ethanol (AE) extracts organic solution.

D. cohinchinensis belong to family of Leguminosae and mainly cultivated in Thailand, Laos and Cambodia, etc. There are abundant flavonoids and phenolic compounds in extracts from *D. cohinchinensis* and some of them have been identified and shown in the Fig. 1(Zhu et al., 2021). The literatures have revealed that the flavonoids and phenolic compounds in the wood extracts were determinant in terms of wood durability (Tascioglu et al., 2013; Windeisen et al., 2002). Accordingly, some wood extracts were employed to investigation in respected to improving the mildew resistance property of fast-grown wood and promising findings can be obtained (Laks et al., 1988; Salem et al., 2014). *Poplar* wood veneers are vulnerable to moldy on accounts of its lower density and durability, which may lessen its service time. However, its mildew resistance property may be improved during the dyeing process in company with desired color appearance obtained attributed to wood extracts served as ND. Normally, dyed wood are also prone to discoloration when encounter the water directly or placing in humid environment (Wang et al., 2018a). Thereby, evaluation of water-resistance and mildew resistance property of the dyed wood exhibits quite necessary addressing multiple functionalization of this ND dyed wood.

In continuation of our previous study, this paper was mainly focused on the evaluation of the wash fastness property of the ND dyed wood by comparison with that of the dyed wood with chemical dyes pertain to the variation of colorimetric value before and after hot-water immersing. And the ND dyed wood against *Aspergillus niger* and *Trichoderma viride* was examined for its durability in respected to the mildew resistance property. Additionally, the exploration pertain to in-depth dyeing process of the ND was also conducted on the wood blocks under atmospheric pressure.

2. Materials And Methods

2.1 Materials

The rotary-cut sapwood samples of *Poplar tomentosa* with no visual flaws including crack, knot and discoloration, etc., collected in Fujian province, were prepared with two kinds of dimensions, *i.e*. 25 × 1.2 ×

60 mm and 25 × 6.0× 60 mm (tangential×radial×longitudinal) and air-dried to make their moisture content less than 8%. The *D. cohinchinensis* residue was cleaned and air-dried in atmospheric environment. Afterwards, it was ground into wood powder and 40–60 mesh was chosen for ND preparation. The chemical dyes (acid red GR:$C_{22}H_{14}N_4Na_2O_7S_2$ and reactive red 3G: $C_{52}H_{34}Cl_4N_{14}Na_7O_{10}S_2$) were purchased from the Jia Ying Chemical Company, in Shanghai, China. And *the Aspergillus niger* and *Trichoderma viride* were supplied by Chinese academy of forestry sciences.

2.2 ND preparation

The *D. cohinchinensis* powder was extracted with anhydrous ethanol (AE) using soxhlet apparatus in the thermostat water bath setting the temperature as 79°C (boiling point of AE) and the organic extractive solution was employed to dye samples directly (Fig. 2).

2.3 Dyed wood preparation

2.3.1 ND dyed wood preparation

The ND dyed wood (both the veneers and blocks) can be obtained by employing extracts from *D. cohinchinensis* as ND under the atmospheric pressure along with the dip-dyeing method reported in the previous study (Liu et al., 2015c). The optimized experimental parameters including dyeing time, dyeing temperature and dyeing concentration were set as 6h, 79°C and 15g/250mL ($m_{wood\ powder}$: V_{AE}) reported in our previous work, respectively (Zhu et al., 2021). The assay process of ND dyed wood was summarized in Fig. 3.

2.2.2 AGR and R3G dyed wood preparation

Based on the reported references (Hu et al., 2016; Liu et al., 2015b), the AGR and R3G dyed wood samples can be obtained by keeping dye temperature 80°C for 4h (bath ratio,1:20) with the dye concentration as 0.5% (w/v). And the mordant 0.5% (w/v) Na_2SO_4 was added in the water-soluble dye solution at the initial (for AGR dyed wood) and ultimate stage of the test (for R3G dyed wood) along with the pH value adjusted to 4–5 by the 10% H_2SO_4.

2.4 Water fastness test of ND dyed wood

Since no standards are available for testing the water fastness of dyed wood accompany with the universal application of AGR and R3G in the wood dyeing study (Hu et al., 2016; Liu et al., 2015b; Wang et al., 2018), the water fastness property of ND dyed wood was evaluated by comparison the total color difference reflectance curves change with that of AGR and R3G dyed wood after immersing in the hot water (80°C) for 3h.

2.5 Mildew resistance test of ND dyed wood

The mildew resistance property of ND dyed wood was evaluated according to Chinese standard GB/T 18261 – 2013 (China National Standardization Management Committee, 2013). The mold control effectiveness (MCE) was calculated based on Table 1 and Eq. (1).

$MCE=(1-D_1/D_0)\times100\%$ (1)

Wherein, D_1 represents average infection value (AIV) of the treated samples and D_0 is the AIV of untreated samples.

Table.1 Grade of surface infection value

AIV	Surface infection area
0	No hypha and mildew
1	Infection area less than 25%
2	Infection area between 25%-50%
3	Infection area between 50%-75%
4	Infection area more than 75%

2.6 Permeability test of ND

The in-between dyed wood block was split along the longitudinal direction and the minimum ND immersing depth was recorded (Fig. 4). The dyeing penetration rate (*DPR*) was calculated according to the Fig. 2 and Eq. (2).

$$DPR = \frac{L_1 + L_2}{L} \qquad (2)$$

Where L was the total length of the sample, and the L_1, L_2 was the minimum ND immersing depth from the ends of the sample.

2.7 Color assessment

The colorimetric parameters of the samples including L^*, a^* and b^* were determined by X-Rite colorimetric analysis (color I7, America) with D65 standard illuminant and 10° standard observer. The total color difference ΔE^* of dyed wood after water fastness treatment was calculated from Eq. (3).

$\Delta E^* = [(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2]^{1/2}$ (3)

Wherein, ΔL^*, Δa^* and Δb^* represent the change of dyed wood before and after washed in brightness ranging from white (100) to black (0), redness(+)/greeness(−) and yellowness(+)/blueness(−), respectively.

The total color difference of the dyed samples before and after washed treatment can be classified based on visual perception levels (Table 2).

Table.2

Relationship between total color difference and people visual sensation

ΔE^*	**Classification**
0-0.5	Negligible
0.5–1.5	Slightly perceivable
1.5-3.0	Perceptible
3.0–6.0	Appreciable
6.0–12.0	Very appreciable
>12.0	Beyond very appreciable
Source: Adapted from Duan et al. (2002).	

Kubelka-Munk equation (Eq. (4)) was employed to calculated the color strength (*K/S*) of the dyed wood with the wavelength ranging from 360-750nm.

$$\frac{K}{S} = \frac{(1-R)^2}{2R} \qquad (4)$$

Where the *R*, *K* and *S* are the diffuse reflectance, the absorption and scattering coefficient, respectively.

The color intensity (*I*) of dyed wood before and after washed treatment was recorded by Eq. (5), as described by Štěpánková et al.(2011).

$$I = \sum_{\lambda=360nm}^{750\text{nm}} \frac{K}{S}(\lambda).\Delta\lambda \quad \Delta\lambda=10\text{nm} \qquad (5)$$

The reduction percentage (*RP*) of the intensity in terms of the dyed wood was calculated by Eq. (6).

$$RP = \frac{\mathrm{Ib - Ia}}{Ib} \qquad (6)$$

Wherein, the Ib and Ia represents the intensity of dyed wood before and after washed treatment, respectively.

2.8 Materials characterization

The micromorphology of ND dyed wood before and after mildew infection treatment was observed with scanning electron microscopy (SEM, Nova Nano230, America). The contact angle of untreated samples, AE treated samples and ND dyed wood was recorded with contact angle meter (DSA-30, Germany). The contact angle of different samples was recorded per 80ms interval time within 10s and 60s.

3. Results And Discussion

3.1 Evaluation of water fastness property of the ND dyed wood

3.1.1 Color parameters changes of the dyed wood after washed

The slighter color fading of ND dyed wood occurred by comparison with ARG and R3G dyed wood after water washed treatment as perceived by the naked eyes (Fig. 5). As shown in Fig. 6b, the ΔE^* of ND dyed wood was only 4.58 mainly resulting from slight decrease of parameter b^* after water washed, which was far less than that of AGR dyed wood (Fig. 6a; ΔE^*:32.82) and R3G dyed wood (Fig. 6c; ΔE^*:26.85). Correspondingly, the classification of surface color change was graded to appreciable while top grade was achieved for AGR and R3G dyed wood based on the data classification listed in Table 2. Consequently, there was an indication that the water fastness property of ND dyed wood was appreciable and acceptable directly from people's perception. It was probably ascribed to ample phenols and flavonoid compound consisting of the ND (Zhu et al., 2021), which were hard to dissolve in the water. Moreover, distribution form variation between dye and wood tissues may also account for this phenomenon. As reported in our previous study (Zhu et al., 2018), the ND was formed a film that anchored on the wood tissues leading to greater coverage area, which was quite different from spots distribution form for acid and reactive dye (Liu et al., 2015b; Wang et al., 2018b). Additionally, the AGR and R3G offer more propensity for water dissolution problems and further result in poor water fastness property.

3.1.2 Surface reflectance and *K/S* change of the dyed wood

Normally, greater *R* value signifies reduction of visible light absorbing by dyed wood resulting in pale color-shade. As shown in Fig. 7a and 7e, both the *R* value of AGR and R3G dyed wood after washed increased pronouncedly compared with that of unwashed samples, which indicated that terrible color fading of AGR and R3G dyed wood occurred. However, the *R* curves of ND dyed wood before and after washed were nearly overlap manifesting only marginal color fading occurrence (Fig. 7c).

Based Kubelka-Munk theory, there is a linear relation between *K/S* value and chromophores variation in range of visual light spectra (Chen et al., 2012; Huang et al., 2012). *K/S* spectra variation reveals the surface color change of dyed wood before and after washed. As shown in Fig. 7b and 7d, drastic decline of *K/S* curves were transparent for AGR and R3G dyed wood by comparison with the unwashed samples. Nevertheless, only slighter *K/S* spectra changes retained for the ND dyed wood (Fig. 7f). Moreover, the ND dyed wood in terms of *RP* for *I* value was far lower (14.0%) than that of AGR (76.14%) and R3G dyed wood (66.52%) as data revealed in Table 3. All the aforementioned analysis further elucidated the water fastness of ND dyed wood was preferable.

Table 3.

Color intensity of different dyed samples before (*Ib*) and after (*Ia*) washed

Sample	*Ib*	*Ia*	*RP*
AGR dyed wood	1627.74 ± 38.71*	388.36 ± 12.82	76.14%
G3R dyed wood	1059.18 ± 24.99	354.59 ± 11.52	66.52%
ND dyed wood	1094.27 ± 24.55	941.01 ± 18.51	14.01%
* represents the standard deviation			

3.2 Hydrophobicity evaluation of ND dyed wood

The contact angles (CA) of different test samples were measured in response to evaluating the hydrophobicity of ND dyed wood. As shown in Fig. 8a and 8b, the CA variation between undyed and AE treated wood performed tiny after 10s, which indicated that AE treatment posed no effect on the hydrophobicity of substrates. Interestingly, the CA of ND dyed wood after 10s (Fig. 8c) and even more than 60s (Fig. 8d) was still apparently greater than that of undyed and AE treated wood. It can be deduced that ND can be the determinant for ameliorating the hydrophobicity of wood. Moreover, the dynamic change curves of different samples were revealed in Fig. 8e in respect to further visualized observing the CA change with the time prolonging. It can be pronouncedly seen that the CA of the ND dyed wood was stable in the whole test process while sharply decline of CA occurred in both the undyed and AE treated wood. All the aforementioned analysis demonstrated the ND can prominently improve the hydrophobicity of wood, which can account for the water fastness property improvement of the ND wood.

3.3. Mildew resistance of the ND dyed wood

Sapwood are invariable prone to suffer organism erosion under favorable environment leading to partial damage of the wood surface (Rodrigues et al., 2012; Schultz et al., 2007; Schwarze, 2007). *A. niger* and *T. viride* growth and distribution on different test samples after 30 days were shown in Fig. 9. It was obviously observed that both the undyed and AE treated wood performed no inhibition effect against both *A. niger* and *T. viride* indicative of no improvement on anti-mold property of wood pertain to AE treatment. However, the ND dyed wood still exhibited certain inhibition behavior against the two molds even after 30 days. Consequently, it can be inferred the ND was the determinant for imparting the dyed wood appreciable mold inhibition property.

Based on the Table 1, the AIV curves variation of different test samples were shown in Fig. 10a and 10b. The AIV of the ND dyed wood against the two molds increased with the time prolonging but was remarkable lower than that of control and AE treated wood. This displayed that the ND dyed wood exhibited preferable anti-molds property even after 30 days. According to Eq. (1), the MCE of AE treated and ND dyed wood was calculated and shown in Fig. 10c and 10d. It can be apparently seen that MCE of ND dyed wood was improved against the two molds after dyeing process. And the inhibition effect of ND dyed wood against *T. viride* (54.25%) was superior to that of *A. niger* (27.00%)due to its greater infection effect.

The micrographs of molds distribution on wood tissues were observed to address mildew resistance property of ND dyed wood. As shown in Fig. 11a, the vessel wall of undyed wood was smooth and the mildew were absent. However, numerous molds were available on undyed wood tissues after anti-mold assay (Fig. 11b), which revealed that the *poplar* wood was extremely vulnerable to mold infection. Noticeably, drastic reduction of molds distribution was occurred in respect to tissues of ND dyed wood (Fig. 11e and 11f). It was indicative of considerable anti-molds property of ND dyed wood ascribed to ND film generation anchoring on the *poplar* wood (Fig. 11d). As reported in the references, the phenolic and flavonoid compounds can break the cytoderm and cytomembrane of the molds leading to protein damage and further exhibited appreciable mildew resistance property (Jin, 2019; Kasiri and Safapour, 2014; Silva et al., 2018; Talaro, 2016; Tascioglu et al., 2013). Consequently, the ample phenols and flavonoids presented in ND may account for the mildew resistance amelioration in terms of ND dyed wood.

3.3 Penetrability of ND on wood blocks dyeing

Table.4

Molecular weight of different dyes

Dye	Components	Molecular weight
Chemical dyes	AGR	556
	R3G	1381
Main phenols in the ND	Vanillic acid	168
	Caffeic acid	180
	Ferulic acid	194
Main flavonoid in the ND	Rutin	610
	Myricetin	318
	Kaempferol	286

The penetrability of ND was evaluated on wood blocks (thickness: 6mm) dyeing compared with that of AGR and R3G employing atmospheric pressure impregnation. As shown in the Fig. 12c, the inner of the wood blocks was completely impregnated with ND. Whereas, the penetrability of AGR and R3G was fairly poor along the radial direction (Fig. 12a and 12b). The DPR of ND dyed wood (100%) was far greater than that of AGR (3.33%) and R3G dyed wood (3.03%), which further explicated the ND performed unexpected impregnation effect (Fig. 12d). Migration and permeation of the dye molecules were the key factors to determine the penetrability of the wood dyeing (Duan, 2002). Additionally, the dye molecular weight and physicochemical property of the dyes liquor was also pivotal for wood dyeing in depth (Bao and Hu, 1990). In current study, no pretreatment was conducted on wood blocks before dye and the molecular weight of the components in ND with exception of rutin were much lower than that of AGR and R3G (Table.4). This may be supposed to account for the preferable penetrability of ND on wood blocks. In addition, another tentative explanation was that higher polarity resemblance between the dye liquor and wood substrate can provoke the amelioration of the dye molecular movement and further lead to considerable penetrability of the dye (Deng et al., 2006; Wizi et al., 2018). Unquestionably, the AE as organic dye solution performed closer polarity to wood substrates than the aqueous solution used in AGR and R3G dye process and resulted in greater penetrability of the ND.

Up to date, different pretreatment on wood blocks was indispensable with respect to the accessibility of dyeing in depth using chemical dyes (Cao et al., 2008; Deng et al., 2006; Hu et al., 2015). Interestingly, the wood blocks inner was saturated with the ND without any pretreatment in present study indicative of unexpected penetrability of the ND. However, the process of dyes immigration was fairly complicated and many obstacles are still to be addressed in depth for exploiting this issue.

4. Conclusion

In present study, the water fastness and molds inhibition property of the ND dyed wood was evaluated. The ΔE^* and I of ND dyed wood declined lesser after washed treatment by comparison to that of AGR and R3G dyed wood indicating the water fastness of the ND dyed wood was reasonably appreciable. This may be attributed to the surface hydrophobicity of ND dyed wood improved prominently and ND film generation and depositing on the surface of wood. Concurrently, the ND dyed wood exhibited mildew inhibition property against *T. viride* and *A. niger* ascribed to ample phenols and flavonoid existed in the ND conferring anti-molds to the *poplar* wood veneers. Additionally, the poplar blocks inner can be impregnated with ND using AE extracts as dye solution under atmospheric pressure. Unquestionably, it gives an evidence that this dyeing scenario can achieve tremendously penetrability in depth of wood blocks.

Combined with our pervious study, the multifunctional property of ND dyed wood has been validated in terms of anti-UV, preferable water-fastness, molds growth inhibition in conjunction with unexpected penetrability behavior of the ND on wood blocks dyeing without any mordant adding. Results of this study can perforce provide potential insight for multifunction dyed wood preparation and guarantee the service time prolonged of the ND dyed wood for possible outdoor usage.

Declarations

Declaration of Competing Interest

The authors report no declarations of interest.

Acknowledgements

Financial support of the present work was provided by Research Fund of Fujian Provincial Finance Department (NO. K8115004A) and Science and Technology Innovation of Fujian Agriculture and Forestry University (NO. KF2015008).

Abbreviations

AGR**,** acid red GR; AE, anhydrous ethanol; AIV, average infection value; *I*, color intensity; CA, contact angles; *DPR,* dyeing penetration rate; MCE, mold control effectiveness; ND, natural dye; R3G, reactive red 3G; RP, reduction percentage; SEM, scanning electron microscopy; ΔE^*, total color difference; UV, ultraviolet

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Figures

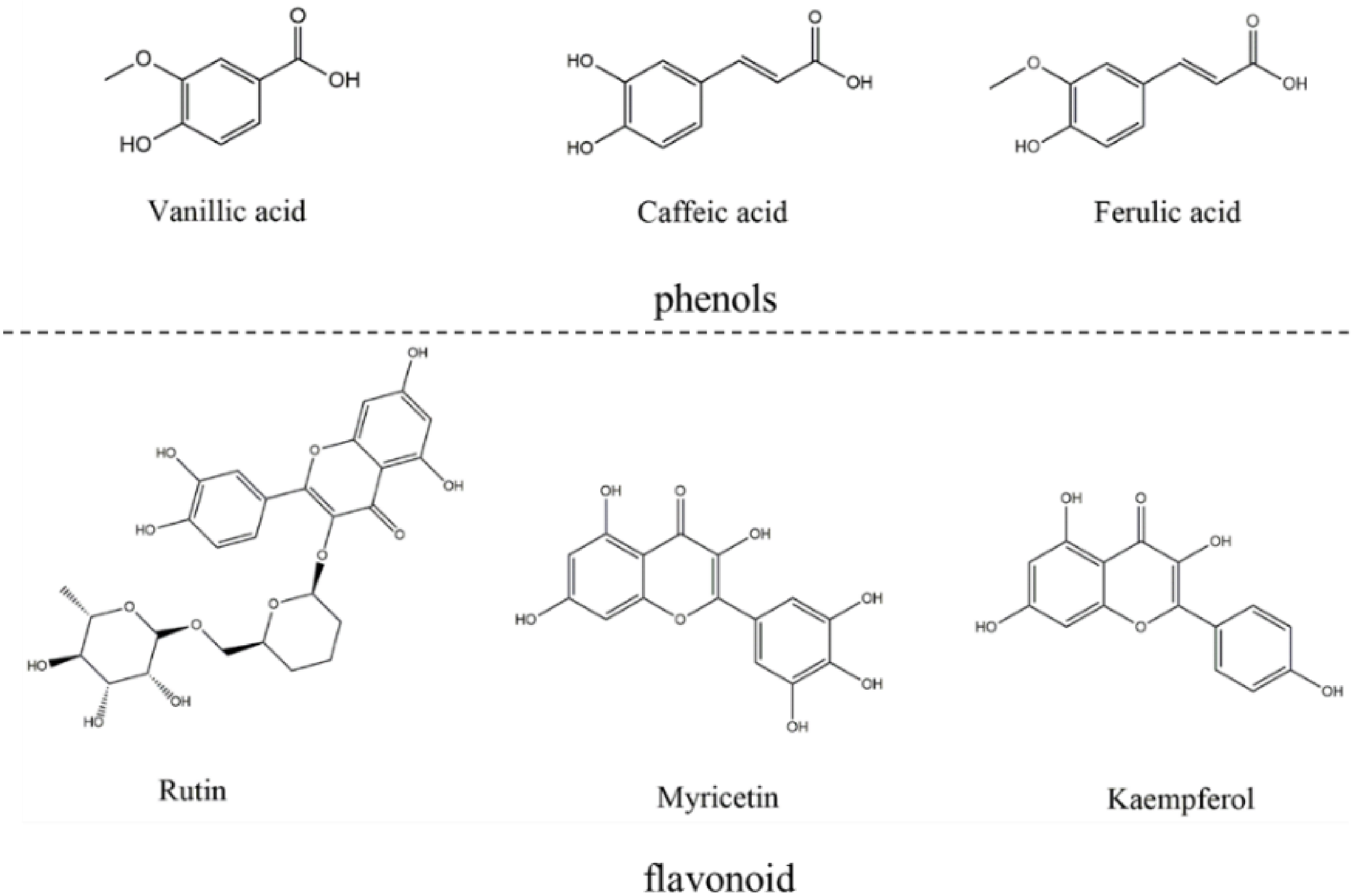

Figure 1

Structures of some main phenols and flavonoid components identified in the ND

Figure 2

D. cochinchinensis residues and extracted ND dye

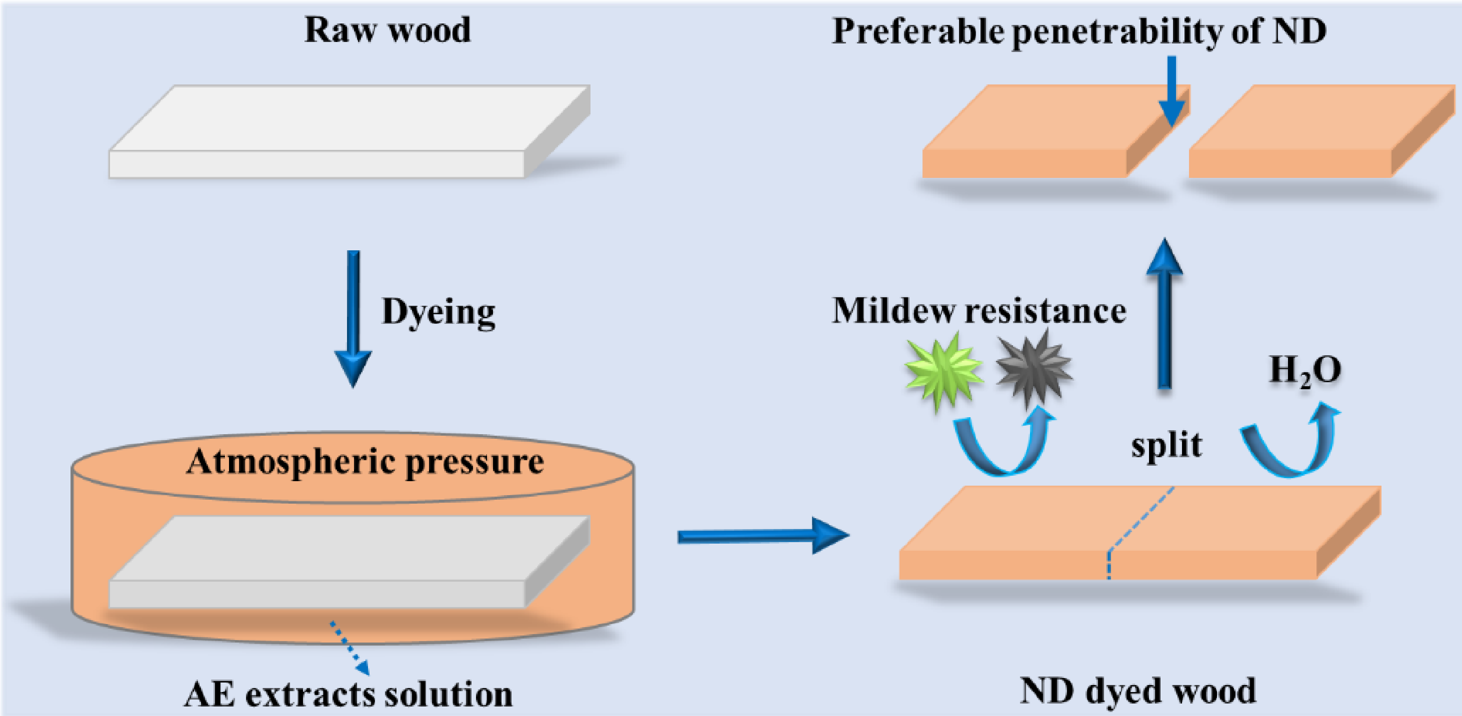

Figure 3

Diagrammatic sketch for multifunctional ND dyed wood preparation

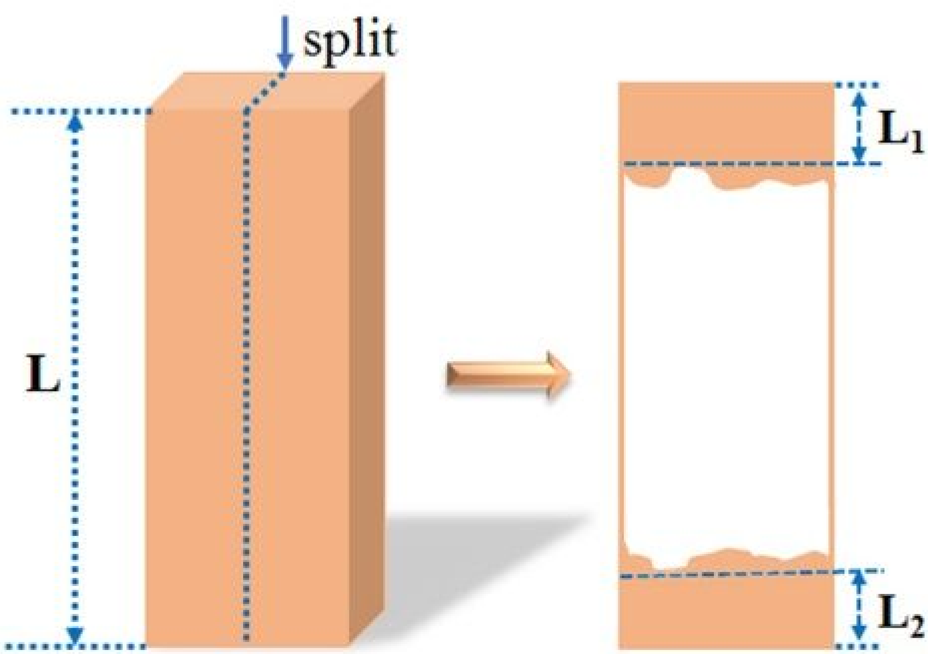

Figure 4

The schematic diagram of calculation of DPR of dyed wood

Figure 5

Photos comparison of ND dyed wood with AGR and R3G dyed wood veneers before and after being washed

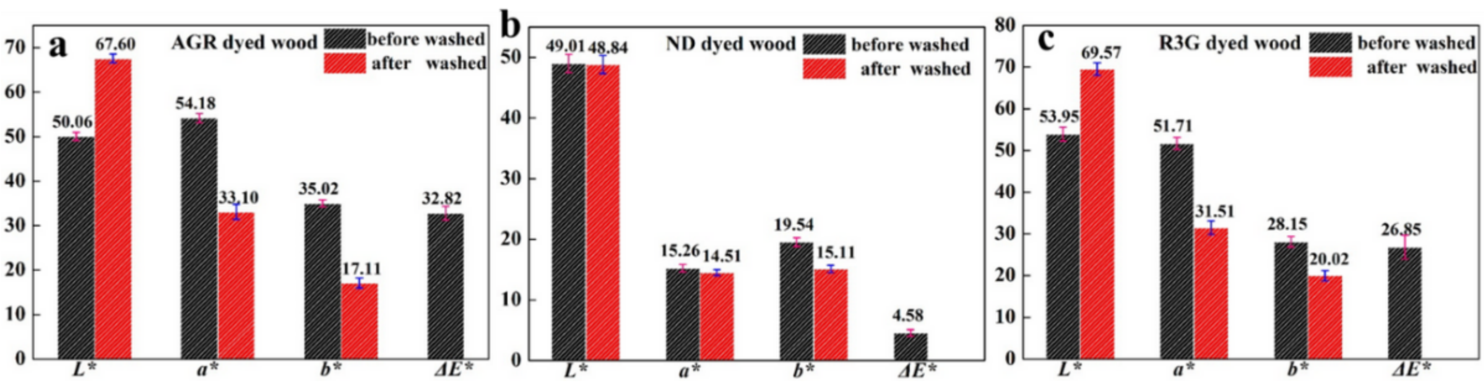

Figure 6

Color parameters and ΔE* change of different dyed samples before and after washed

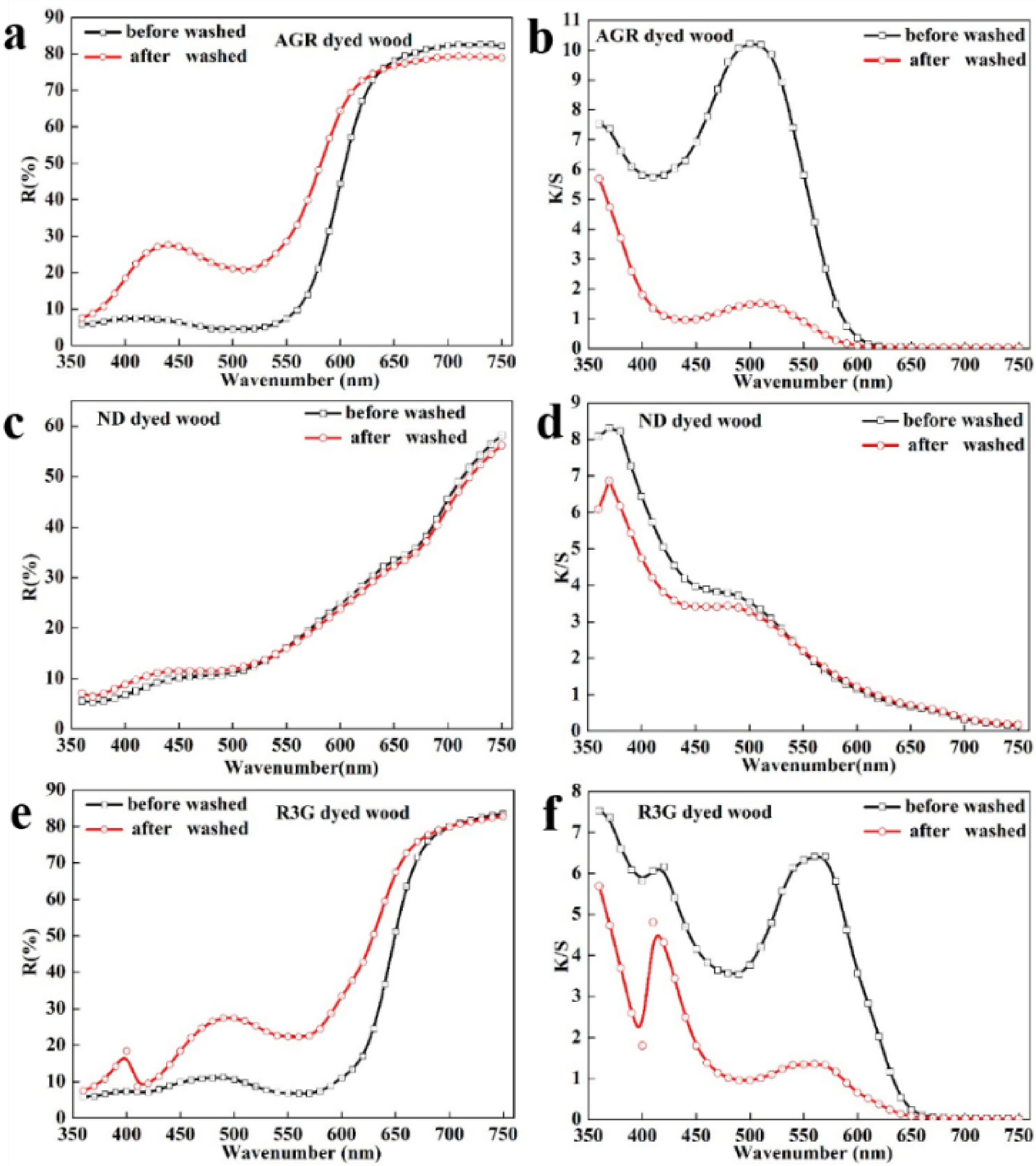

Figure 7

Reflectance and K/S spectra variation of different dyed samples before and after washed

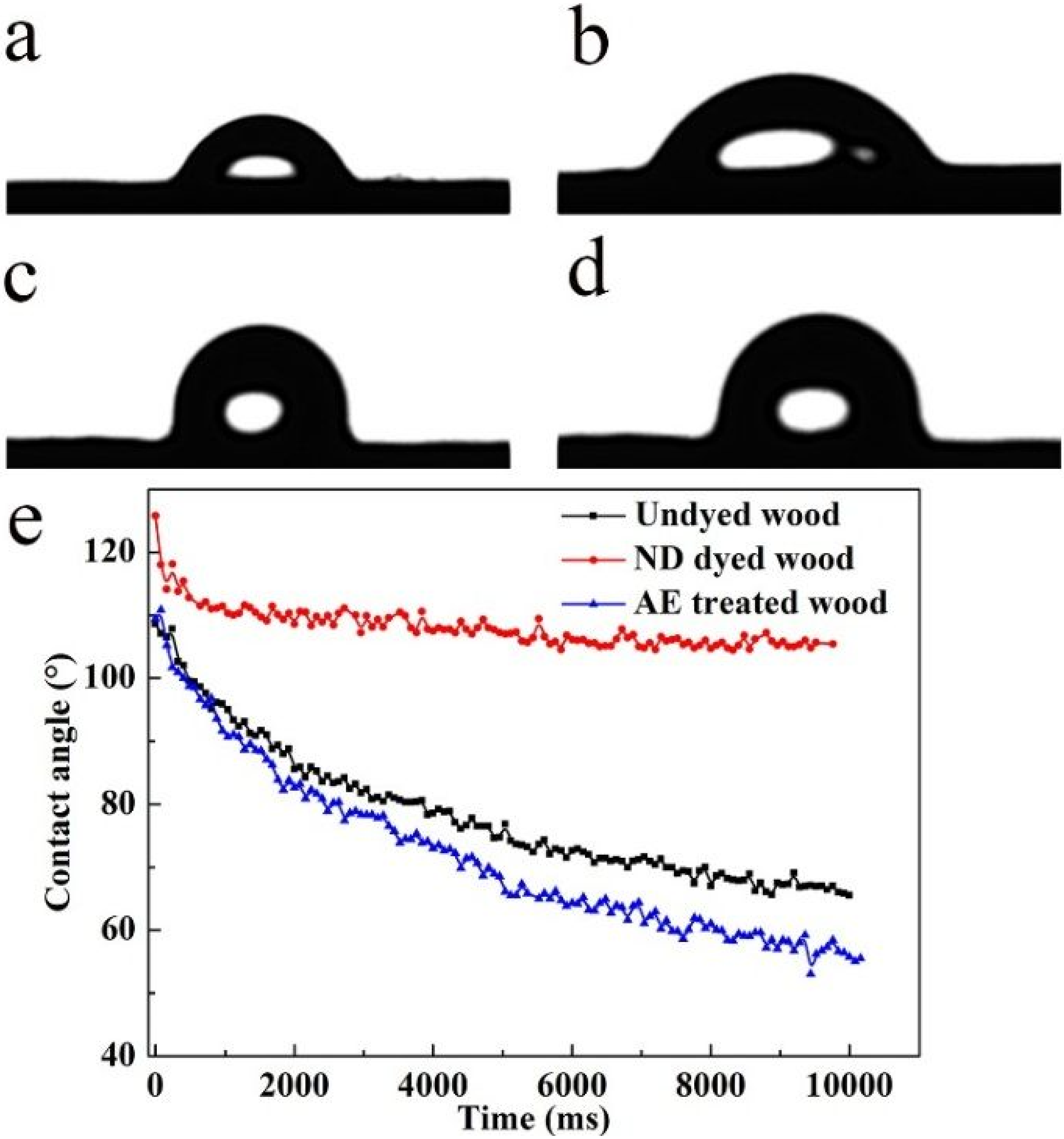

Figure 8

(a): Surface contact angle photo of water on undyed wood after 10s; (b): Surface contact angle photo of water on AE treated wood after 10s; (c): Surface contact angle photo of water on ND dyed wood after 10s; (d): Surface contact angle photo of water on undyed wood after 60s; (e): Surface contact angle change of water on different samples.

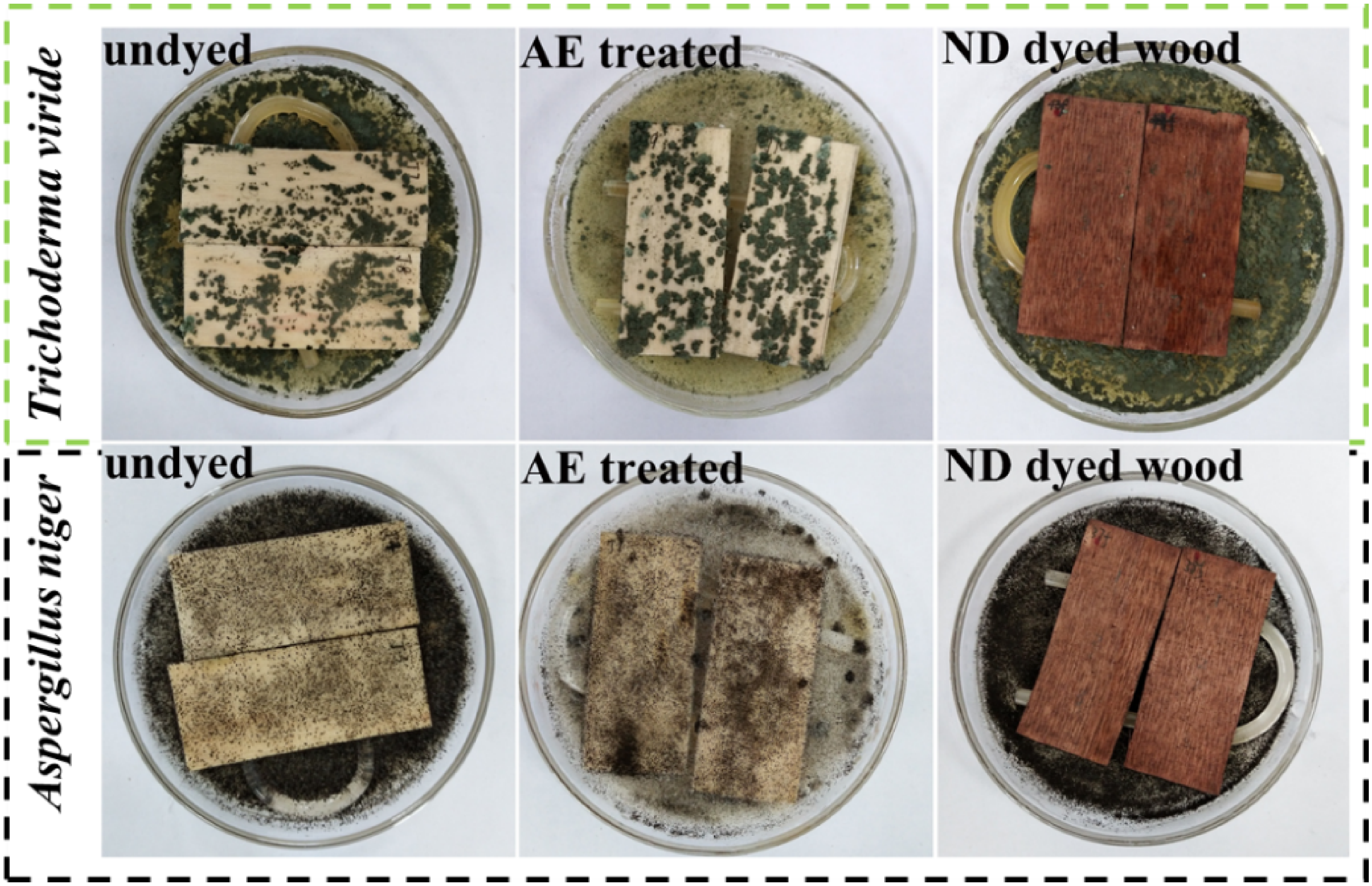

Figure 9

The infection photos of different test samples against A. niger and T. viride after 30days

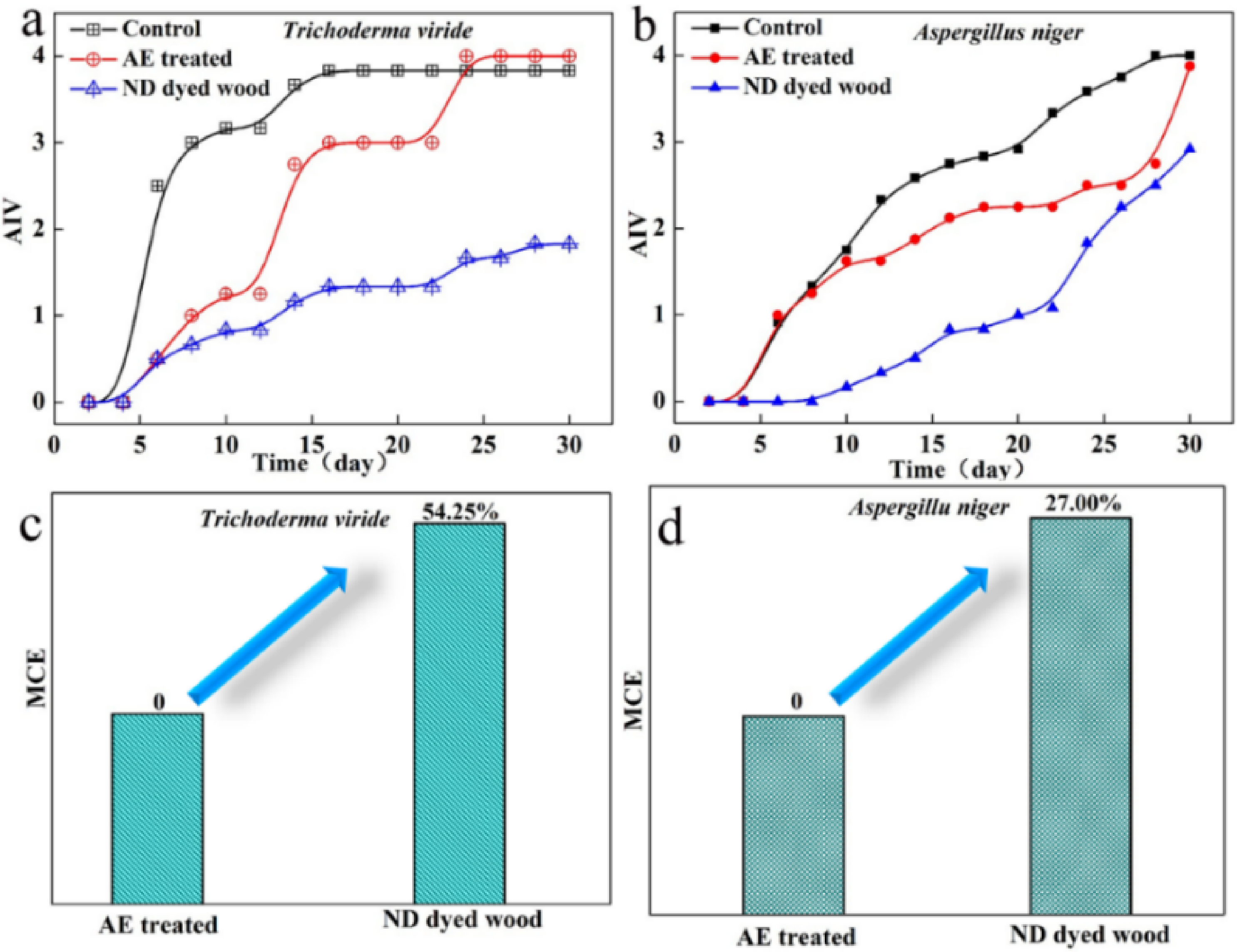

Figure 10

Trends of AIV of different test samples against T. viride (a) and A. niger (b); MCE of ND dyed wood against T. viride (c) and A. niger (d)

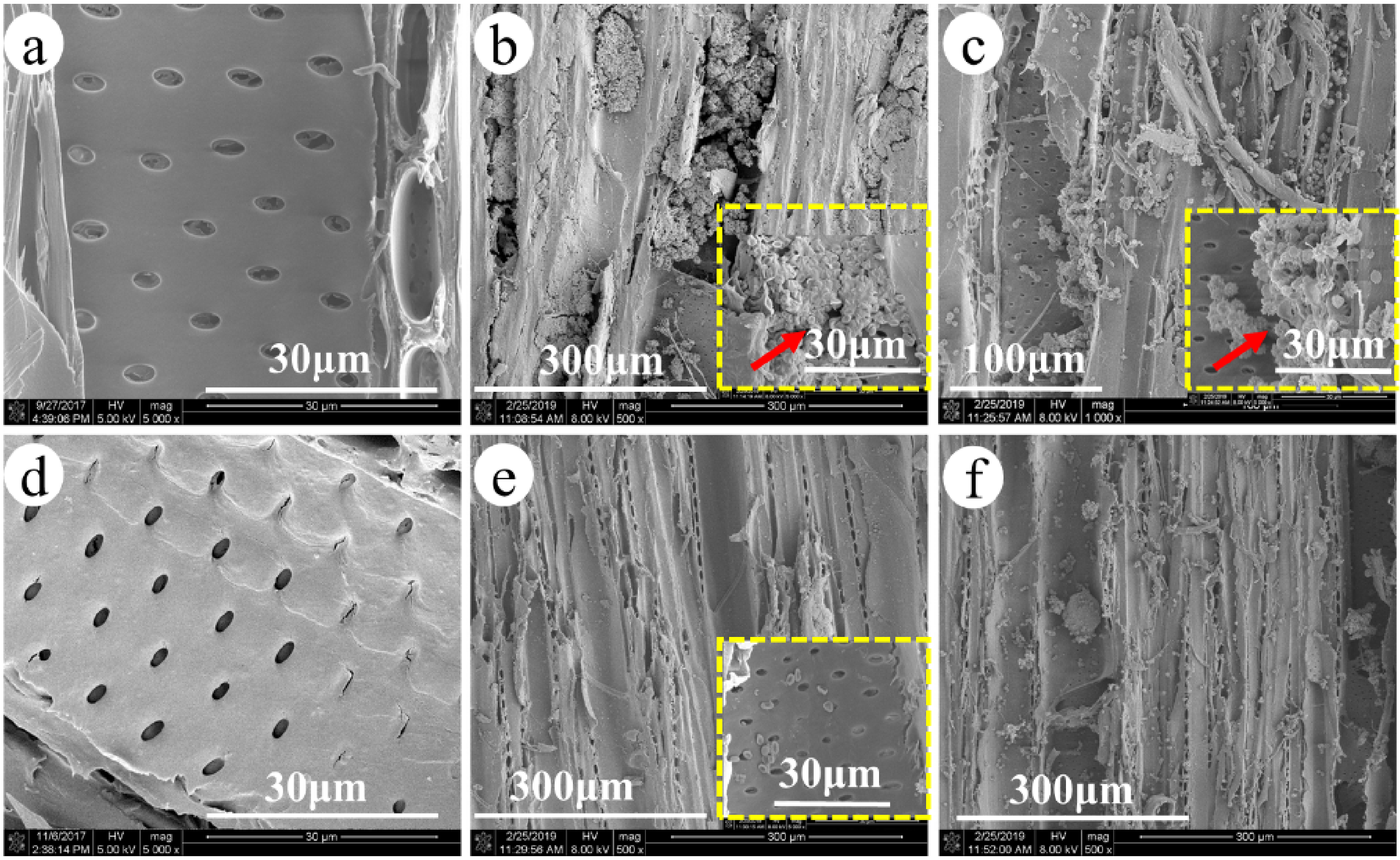

Figure 11

SEM micrographs of undyed wood (a), undyed wood against T. viride (b) and A. niger (c), ND dyed wood before anti-molds test (d), ND dyed wood against T. viride (e) and A. niger (f)

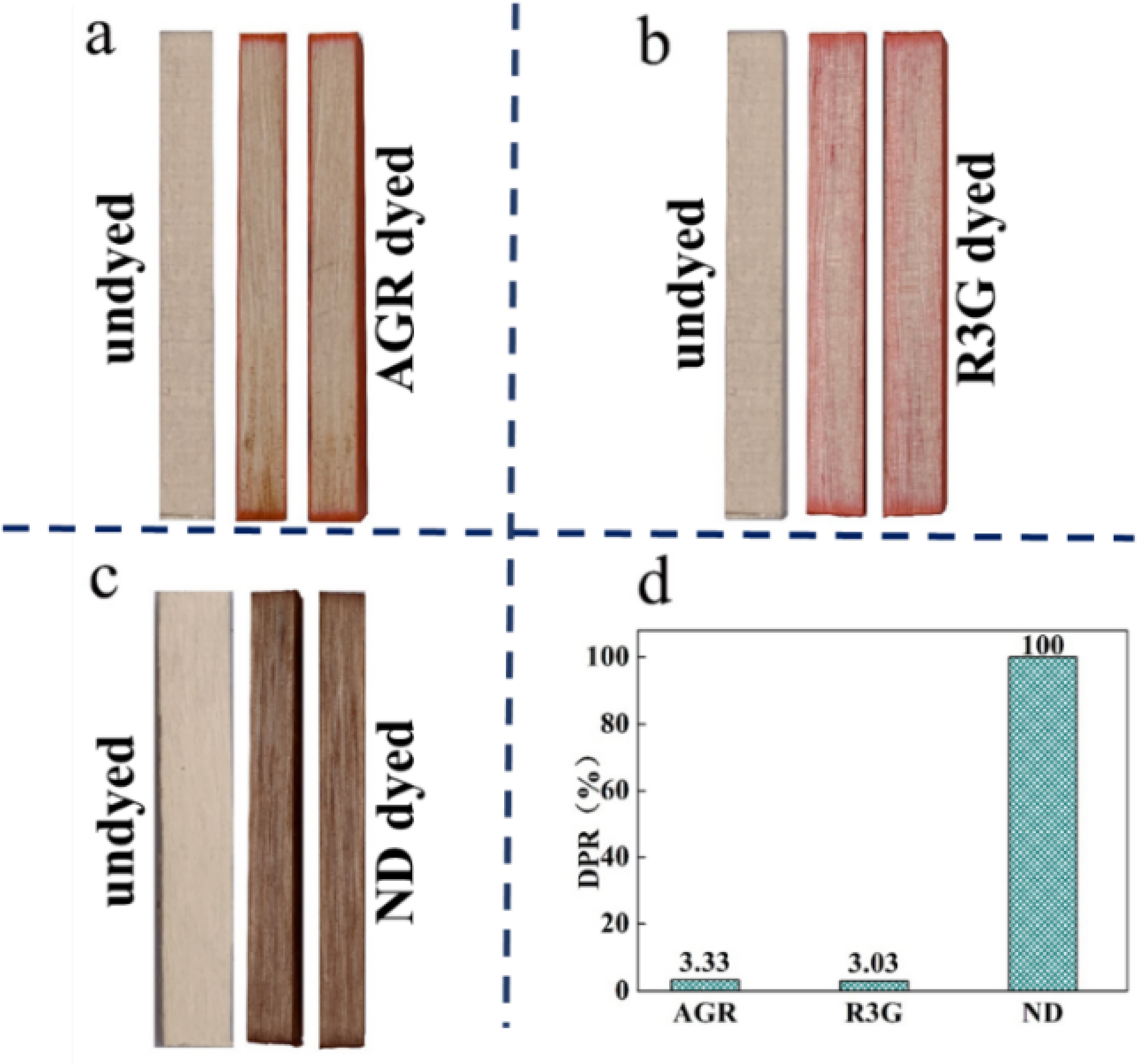

Figure 12

Photos of wood blocks (thickness:6mm) dyed with AGR(a), R3G(b), and ND(c); the DPR of wood blocks dyed with different dyes.