# NOTE

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# Blackening of sugi (*Cryptomeria japonica* D. Don) heartwood in relation to metal content and moisture content\*

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Abstract Blackening in heartwood was investigated in relation to the metal contents and the moisture content in xylem of about 50-year-old seedling sugi (Cryptomeria japonica D. Don) planted in a steeply sloped stand in Okutama district (Itsukaichi Tokyo), where blackened heartwood is frequently found. The potassium, calcium, iron, and manganese contents were examined in the variously blackened heartwood and normal heartwood by an atomic absorption method. It was recognized that potassium increased relative to the degree of the blackening of heartwood, resulting in a significant correlation between them. This finding implies that an increase in potassium has an important role in the blackening of heartwood. Moisture content has a tendency to increase in the blackened heartwood, so it seems that the large accumulation of potassium is associated with the high moisture content in heartwood.

Key words Black-heart · Ash content · Potassium content · Moisture content

### Introduction

In sugi (*Cryptomeria japonica* D. Don) wood it is normal that the heartwood assumes a red-brown to rose-pink color, although it also uncommonly turns black-brown or black. The latter has been called black-heart. The blackening of heartwood is known to occur either in a growing tree or

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after the cutting of trees. The discoloration, or blackening, of wood in a growing tree may be brought about by wounds incurred by pruning or noxious insects, whereas the blackening that occurs after cutting trees may be concerned with metallic components, alkalinity, or some microbe. It has also been reported in sugi that genetics, site, soil, and silvicultural practices are important factors that cause blackheart or affect the extent of blackening in the heartwood.<sup>1-5</sup> In fact, some breeds of sugi frequently result in black-heart. Furthermore, sugi trees of black-heart are more often found at a lower site of sloped stands near the valley than at the upper site. In some cases the black discoloration occurs more often at the upper site near the ridge than at the lower site.

In this study the occurrence and cause of black-heart in sugi trees were investigated in relation to some metal contents and moisture content in the heartwood. The sugi trees used were from Tama district (Itsukaichi, Tokyo) where black-heart is frequently found.

# **Materials and methods**

Approximately 50-year-old sugi trees of seedlings that had been planted over the whole slope from valley to ridge (gradient of about 30 degrees) of Okutama district (Itsukaichi, Tokyo) were used as material. A total of 52 samples were obtained randomly from a lower site in the stand (A plot) near the valley, and 21 were from the midslope in the same stand (B plot) situated 50m up from A plot. Disks 5 cm in thickness were cut 20 cm above the tree base, and blackening was examined in the heartwood of the sections 1h after cutting. The variously blackened disks were grouped into three types by visual inspection: strong, medium, and thin black; four, five, and seven disks were obtained from the three types, respectively. In addition, nine disks were obtained from samples with normal heartwood. Blocks 3-5cm from each of 25 disks were cut successively from sapwood to heartwood, exclusive of intermediate wood. A total of 98 samples were obtained to mea-

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Examination sites in the stand <sup>a</sup>	No. of samples	Frequency of black-heart			
		Degree of blackening	No.	Total	
A plot	52	Strong	4 (7.7%)		
		Medium	5 (9.6%)	20 (38.5%)	
		Thin	11 (21.2%) <sup>J</sup>		
B plot	21	Strong	0		
		Medium	1 (4.8%)	3 (14.3%)	
		Thin	2 (9.5%)	. ,	

Table 1. Frequency of occurrence of black-heart in a sloped stand of sugi

<sup>a</sup>A plot is a lower site in the sloped stand, and B plot is a mid-slope in the same stand.

sure the metallic and moisture contents in sapwood and heartwood.

Blocks were weighed immediately after cutting from disks and again after drying for 1 day at  $105^{\circ}$ C to determine the moisture content in the sapwood and heartwood. The brightness (L\*) in the cross section of the blocks, left for a few days after cutting, was measured with a colorimeter (Suga shikenki H-CT) to evaluate the extent of the blackening in the heartwood.<sup>6</sup>

Potassium, calcium, iron, and manganese contents were analyzed by an atomic absorption method. Blocks of sapwood and heartwood were burned in a mafful furnace at 450°C for 30min, then at 700°C for 40min, and finally at 850°C for 2h. The ashes were dried for measurement of the true weights and were dissolved in dilute 6N hydrochloric acid for analysis of the content of each metal in sapwood and heartwood with a spectrophotometer (Hitachi 170-30 type).



## **Results and discussion**

Occurrence of black heartwood in the stand

The color of heartwood was normal in the sections just after cutting. Within 1 h, however, the heartwood became thin or dark black (normal color is red-brown). Such blackened heartwood, also called black-heart, was grouped into three types according to the degree of blackening by visual inspection: strong black (BH-I), medium black (BH-II), and thin black (BH-III). Occurrences of black-heart in each type differed with the position of the tree in the stand, as shown in Table 1. The frequency was 40% among trees from the lower site of the sloped stand near the valley, which was more than twice that in trees from the upper site. In addition, the type evaluated as strong black was not recognized in trees from the upper site.

It has been reported that blackening in the cut sections, which occurs with the length of time after cutting, may be brought about by oxidation of metals (e.g., iron and manganese) or alkalization of water in the heartwood.<sup>7-10</sup> It is suspected that iron and manganese give rise to the blackening by chelate linkage to a catechol substance in sugi heartwood.

Fig. 1. Average contents of ash in sapwoods and heartwoods of blackheart and red-heart. *BH*, black-heart; *BH-I*, strong black heartwood; *BH-II*, medium black heartwood; *BH-III*, thin black heartwood; *RH*, red-heart

Another possible cause is oxidation of peculiar phenol substances contained in the heartwood of sugi on exposure to air, resulting in blackening of the heartwood. The oxidizing reaction of the phenol substances for blackening of heartwood progresses under a weak alkaline condition, which may be related to the increase in alkaline metals, (e.g., potassium). Therefore it is of profound interest to know the content of potassium, iron, and manganese in heartwood in relation to the blackening. Finally, it was suspected that these contents are associated with the frequency of occurrence of black-heart in the sugi stand.

#### Content of ash and metals

Figure 1 shows that in red-heart the content of ash in the heartwood was larger than that in sapwood. Although the tendency was the same in the xylem of blackened heartwood, the ash content in the heartwood was larger than that in red-heart, and the content increased as the degree of blackening became greater. The difference seen between black-heart and red-heart in the heartwood was not recognized in sapwood. The increased ash content corresponding with the blackening of heartwood was closely associated with the potassium and calcium contents, as shown in Table 2. These correlation coefficients were statistically significant at the 1% level. The high correlation coefficient between ash content and potassium content in heartwood is particularly emphasized in Fig. 2. This result coincides with those in other reports.<sup>11</sup> As shown in Table 3, the increase in potassium content corresponded thoroughly with the degree of the blackening; in the most blackened heartwood the content showed an increase of about 70% compared to that of normal heartwood. This tendency was not seen for the other metals or in sapwood. Iron and manganese contents, which might bring about formation of a complex salt of dark color by the reaction with phenolic substances in heartwood, did not increase characteristically in the blackened heartwood. This result implies that the formation of blackened heartwood is hardly related to these metals.

The marked increase in potassium content indicates a close relation to L\*, which represents the degree of blackening of heartwood as measured by colorimetry. The correlation coefficient is 0.678, which is significant at the 1% level (Table 2). Although the iron content is significant at the 5% level in relation to L\*, it is difficult to conclude that the substance brings about the blackening of heartwood, because the coefficient value is relatively low compared to the potassium content. In addition, Kai<sup>7</sup> has reported negative

Table 2. Correlation coefficient between ash content, L\*, or moisture content and each metal content in heartwood of black-heart and red-heart

Metallic elements	Coefficient					
	Ash content (%)	L*	Moisture content (%)			
K	0.858*	0.678*	0.634*			
Са	0.315*	0.027	0.056			
Fe	0.176	0.286*	0.199			
Mn	0.108	0.180	0.148			

L\*, brightness; \* Significant at 1% level; \*\* significant at 5% level.

results regarding the participation of these metals in the blackening of heartwood. Calcium, contained largely in heartwood next to potassium, was not significantly concerned with the blackening.

It is supposed that a high content of potassium in heartwood gives rise to the formation of an alkali salt (potassium carbonate or potassium hydroxide), and as a result, the water in the heartwood is alkaline. It has been proved by Abe et al.<sup>9,10</sup> that the phenolic substance in heartwood is easily blackened by oxidation under alkaline conditions. In addition, in a recent report by Takahashi<sup>12</sup> it has been concluded that the black coloration of sugi heartwood is related to a change in the norlignans in weak alkaline condition with potassium hydorogen carbonate. Therefore it is believed that the increase of potassium content plays an important role in the blackening of heartwood.

The high frequency of black-heart that occurs in trees from the lower site of the sloped stand, near the mountain



Fig. 2. Relation between potassium content and ash content in heartwood of black-heart and red-heart

 Table 3. Metal contents in the sapwoods and heartwoods of black-heart and red-heart

Type of heartwood	Sapwood (mmol/kg)			Heartwood (mmol/kg)				
	K	Са	Fe	Mn	K	Са	Fe	Mn
BH-I	16.6	24.0	0.028	0.026	87.4	24.4	0.030	0.017
BH-II	16.6 (3.1)	(2.0) 22.5 (3.5)	(0.052) (0.026) (0.031)	(0.009) (0.034)	(0.0) 75.1 (11.2)	(0.0) 23.7 (9.5)	(0.010) (0.078) (0.065)	(0.005) 0.022 (0.015)
BH-III	19.2 (6.9)	(5.0) (4.0)	(0.028) (0.040)	(0.005) (0.031) (0.009)	69.6 (12.2)	27.7 (10.9)	(0.005) (0.048) (0.044)	(0.013) (0.018) (0.004)
RH	15.1 (5.8)	25.3 (3.7)	0.021 (0.051)	0.038 (0.024)	51.1 (8.3)	24.1 (8.1)	0.043 (0.032)	0.020 (0.023)

See Fig. 1 for explanation of BH-I, BH-II, BH-III, and RH. Numbers in parentheses are the SD.



Fig. 3. Average moisture content in the sapwoods and heartwoods of black-heart and red-heart. See Fig. 1 for explanation of abbreviations

stream, may be related to the potassium content or moisture content in the soil. For example, if the soil contains a large amount of potassium and its moisture content is high, trees would be able to absorb efficiently the potassium ion dissolved in the moisture in the soil. This particular phenomenon was not investigated in the present experiment. It is also of particular interest whether the ion absorbed from roots can transfer to heartwood. In coniferous species the moisture content in sapwood is more than 200%, whereas in heartwood it usually decreases to 50%-60% with heartwood formation. It is also generally known that bordered pits of tracheid in heartwood are almost closed by torus. However, it has been pointed out that the greater part of the tracheid pit pairs are partially opened or loosely closed in wetwood of todomatsu (Abies sachalinensis).<sup>13</sup> If the bordered pits are not closed completely, and consequently water can move in the heartwood, the moisture content and metallic elements such as potassium increase in the xylem.

#### Relation between metal contents and moisture content

As mentioned in a few reports, the higher moisture content in heartwood is a characteristic of blackened or discolored wood,<sup>4,14</sup> a phenomenon confirmed in this study (Fig. 3). The moisture contents in blackened heartwoods were above 100%, whereas those in normal heartwood were about 70%. Furthermore, the correlation coefficient between moisture content and L\* in heartwood, which is 0.669, is statistically significant, suggesting that increases in moisture content relate to the blackening of heartwood.

The metallic ions dissolved by water in soil (e.g., potassium) are carried through the root and sapwood to the leaves or other tissues in the stem by transpiration. Then, if the bordered pits of tracheids in heartwood are not completely closed for any reason, potassium ions may be able to move in the xylem, and the substance accumulates in the heartwood. In fact, it appears that an increase in moisture content in heartwood is associated with an increase in metals, especially potassium (Table 2). Consequently, the conspicuous increase in potassium associated with an increase of moisture content may play a principal role in the blackening of heartwood.

The question arises why potassium alone increases conspicuously in the blackened heartwood corresponding with the increase of moisture content. The answer may be that the mobility of potassium ions is higher in heartwood than other metals, resulting in a high potassium content in this region, although confirmation of this explanation has not been attempted.

# Conclusions

Black-heart of sugi occurred frequently in trees from the lower site of a sloped sugi plantation near a mountain stream. A marked increase in ash was recognized in the blackened heartwood, with the amount being about twice that of normal heartwood, which has a red-brown or a rose-pink color. In particular, the potassium content increased about 70%, whereas other metals (e.g., calcium, iron, and manganese) did not quantitatively increase in the black-ened heartwood. This finding has been confirmed by other workers.<sup>11,15</sup> Therefore it is concluded that the peculiar increase in potassium in the blackened heartwood is evidence that it takes part in the blackening process. This conclusion coincides with suggestions of Abe et al.<sup>9</sup> and Takahashi<sup>12</sup> that potassium plays an important role in the formation of blackened heartwood.

It appears that the increase of potassium corresponding with the blackening of heartwood is closely associated with the distribution of moisture content in the stem of a tree. A higher moisture content in the heartwood enables the potassium ions to move into the heartwood. In fact, the moisture content is high in blackened heartwood - more than twice that of with normal heartwood - and the correlation between the potassium content and the moisture content is significant. The increased moisture content in heartwood may be explained by the permeability of water from branch stubs<sup>16</sup> and osmotic potential.<sup>13</sup> Simultaneously, the opening of bordered pits of intertracheid in heartwood must be associated with the high moisture content. These proposals and mechanisms have not been confirmed, and the cause that alkaline metals (e.g., potassium) increase blackened heartwood has not been examined.

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## References

- Nakata G, Sugawara I, Migita K (1986) Relationship between soil fertility and heartwood color of Sanbusugi (*Cryptomeria japonica*) (in Japanese). Trans Jpn For Soc 97:345–346
- 2. Nakata G, Sugawara I, Migita K (1987) The heartwood color of sanbu-sugi (*Cryptomeria*) (in Japanese). Jpn For Tree Breed 145:1-4
- Watanabe M (1977) Examination on heartwood color of sugi (Cryptomeria japonica) (in Japanese). Jpn For Tree Breed 105:21– 24
- Miwa Y (1991) Moisture content in green wood (in Japanese). In: Mokuzai no Kagaku to Riyo Gijutsu. II. Sugi. Report of a Sectional Committee of the Japan Wood Research Society, pp 10–13
- Mishiro A (1991) Wood color (in Japanese). In: Mokuzai no Kagaku to Riyo Gijutsu. II. Sugi, Report of a Sectional Committee of in Japan Wood Research Society, pp 31–33
- Iwagami M, Hashimoto N (1980) On color variation in heartwood of sugi (in Japanese). Bull Kochi Univ For 7:117–123
- Kai U (1975) Color of wood (1) (in Japanese). Wood Ind 30:291– 294
- 8. Takeuchi S (1986) Stains of sugi (*Cryptomeria japonica*) wood treated with alkali salt solutions (in Japanese). Trans Jpn For Soc 97:351-352
- 9. Abe Z, Oda K, Matsumura J (1994) The color change of sugi (*Cryptomeria japonica*) heartwood from reddish brown to black. I.

The color changes and its causes (in Japanese). Mokuzai Gakkaishi 40:998–1005

- Abe Z, Oda K, Matsumura J (1994) The color change of sugi (Cryptomeria japonica) heartwood from reddish brown to black. II. Identification of potassium hydrogen carbonate as one of the causative materials (in Japanese). Mokuzai Gakkaishi, 40:1119– 1125
- Morikawa T, Oda K, Matsumura J, Tsutsumi J (1996) Black heartwood formation and ash content in the stem of sugi (*Cryptomeria japonica* D. Don). II. Heartwood properties of three sugi cultivars (in Japanese). Bull Kyushu Univ For 74:41–49
- Takahashi K (1996) Relationships between the blackening phenomenon and norlignans of sugi (*Cryptomeria japonica* D. Don) heartwood. I. A case of partially black-heartwood (in Japanese). Mokuzai Gakkaishi, 42:1119–1125
- Ishii T, Fukazawa K (1987) Sap constituents and pit closures connected with water penetration in the wetwood of *Abies sachalinensis* Masters (in Japanese). Res Bull Hokkaido Univ For 44:1277-1305
- Fujiwara S, Iwagami S (1989) Relationship between green moisture content and heartwood color of sugi and hinoki (in Japanses). Bull Kochi Univ For 16:19–23
- Oda K, Matsumura J, Tsutsumi J, Abe Z (1994) Black-heartwood formation and ash contents in the stem of sugi (*Cryptomeria japonica* D. Don) (in Japanese). Bull Fac Agric Kyushu Univ 48:171–176
- Bauch J, Holl W, Endeward R (1975) Some aspects of wetwood formation in fir. Holzforschung 29:198–205