

STUDIES OF JAPANESE OLD TIMBERS

By

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Summary

This paper presents a survey of our studies on the permanence of wood, which have elucidated the outline of changes in the physical, mechanical, and chemical properties of those timbers for building which have stood the test of time for many centuries.

Emphasis is placed on the resemblance between the processes at a room temperature and those of a high temperature of about 100°C, and on the difference between two kinds of wood, Hinoki, a soft wood, and Keyaki, a hard wood.

The obtained conclusions and suggestions are as follows :

1) The cellulosic materials in the timber show a variation with time not only in their quantity but also in their quality. Their content decreases in these two kinds of wood, and their chemical inertness, observed in their crystalline-region content and in their viscosity, increases markedly at the beginning of the ageing and the

heating of Hinoki.

2) These variations may be the main factors of the changes in the properties of timbers in consideration, such as shrinkage, hygroscopicity, strength properties, and content of the soluble constituents. The estimated rate of decrease in the cellulosic materials and the rates of other changes offer a parallel case through a variety of conditions, and the increase in their inertness accompanies with increase in some kinds of mechanical strength.

3) The content of the cellulosic materials in Keyaki decreases faster than that of Hinoki, and the initially superior strength of the former drops down below that of the latter within several centuries. This difference in the permanence between the timber of Hinoki and that of Keyaki is expected to represent the general difference between soft woods and hard woods.

I. INTRODUCTION

Few works on the permanence of timber have been done, partly because they must treat very little change with time of its properties while there is great variance in their intrinsic values, and partly because their suitable materials are not so widely distributed in the world.

Now, in this country, many of ancient wooden edifices have been preserved, among which we have the oldest wooden building in the world—the HORYU-JI Temple, erected in the seventh century. These old buildings provide precious sources of our testing material, preserved under favourable conditions, and about

150 members of these old timbers have been collected by us.

Among these members, nine species of wood are found; Hinoki, *Chamaecyparis obtusa* ENDLICHER, covers about a half of them, and Keyaki, *Zelkova serrata* MAKINO, is next in frequency. The former species, a soft wood, and the latter, a hard wood, have been regarded as two representatives of wooden building-materials in this country.

This paper deals with the following properties of these two species of timber, free from any defect :

Shrinkage,

Hygroscopicity,
 Mechanical properties,
 Chemical composition,
 Colour,
 Crystalline-region content of cellulose,
 Viscosity of cellulose in cuprammonium.

In addition to these old timbers, thirty specimens of unearthed woods which had been buried for thousands of years have been collected and analysed. They are thought to indicate the extreme of ageing.

During the course of conducting these investigations, it was expected that the natural transitions found in old timbers and unearthed woods in consideration, are promoted under some artificial conditions. One of these conditions has been obtained by the heating of wood at 100° ~130°C, far below the temperature of its carbonization.

The results of the investigations are given in the following thirteen figures, and it is easy to compare the hard wood with the soft wood, and the heat-treated wood with the old timbers, in these figures.

II. EXPERIMENT

The members of the old timbers have been obtained, with the judgement of their age by experts in the history of architecture. Their portions, which were examined to have no defect by the naked eye and were proved to be free from wood-destroying fungi, have been used for the investigations. To these, some new specimens have been added for comparison. The ages and sources of the old timbers are given in Table 1. The scarceness of Keyaki before the fourteenth century is believed to be due to the infantile manipulation of the hard wood in those days.

The descriptions of some specimens of the unearthed wood will be given in paragraph 2-5.

For the determination of the strength properties the standard method of J.E.S. has been followed. The procedures of the chemical analysis have been the same as the standard method of Government Forest Experiment Station in

Table 1. The age and source of the old timbers

HINOKI (<i>Chamaecyparis obtusa</i> ENDLICHER)		
Age	Source	
1,300	HORYU-JI,	NARA Prefecture
1,200	GOKURAKU-IN,	NARA City
900	BYODO-IN,	UJI City
730	DAIHO'ON-JI,	KYOTO City
700	RENGEO-IN,	KYOTO City
530	TOMYO-JI,	KYOTO Prefecture
350	KODAI-JI,	KYOTO City
350	EM'MAN-IN,	OTSU City
350	TO-JI,	KYOTO City
300	NIN'NA-JI,	KYOTO City
KEYAKI (<i>Zerkowa serrata</i> MAKINO)		
650	GOKURAKU-IN,	NARA City
530	TOMYO-JI,	KYOTO Prefecture
350	HOKKE-JI,	NARA City
350	KODAI-JI,	KYOTO City
350	NIJO-JO (Castle)	KYOTO City
320	KIYOMIZU-DERA,	KYOTO City
310	ENRYAKU-JI,	OTSU City
240	KAN'NON-JI	KYOTO Prefecture

Note: For the investigations of colour, specimens are also obtained from other sources not cited above.

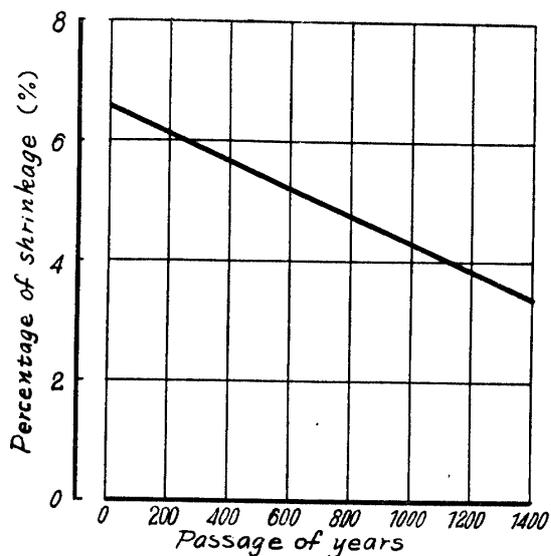
Japan, except that the determination of the cellulosic materials has followed the method of WISE. The procedures to determine other quantities will be given briefly under the following headings.

For the investigation of heat-treatment, the new wood has been heated chiefly in saw dust or in test pieces. It was subjected to oven drying or to heating in a sealed glass-tube.

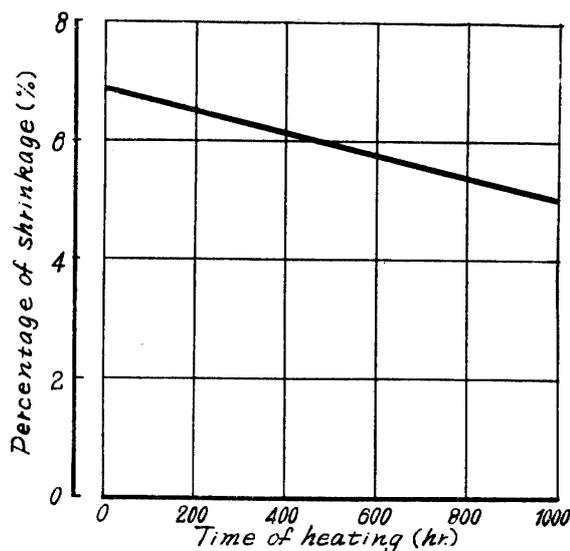
2-1. Shrinkage ⁹⁾

Fig. 1 A gives the relation between the shrinkage and the age of the timber. The tangential shrinkage decreases with the age of the timber.

The test pieces sized 30×30×1.2mm were repeatedly dried with phosphoric acid anhydride and moistened with saturated water vapour, each process taking a week at a constant temperature of 45°C. The shrinkages and swellings were

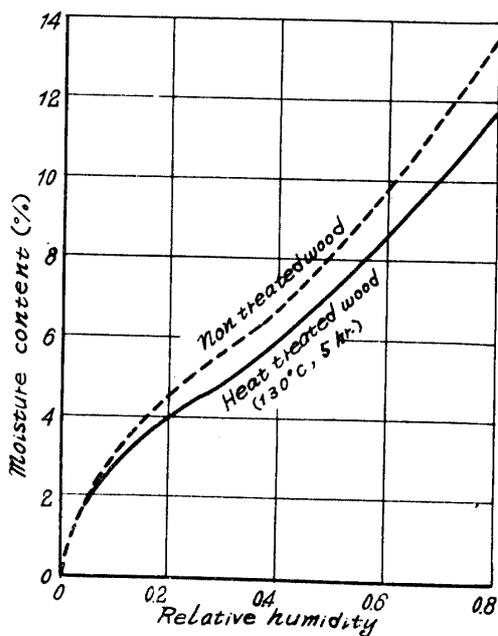


A Old timber (HINOKI, tangential)

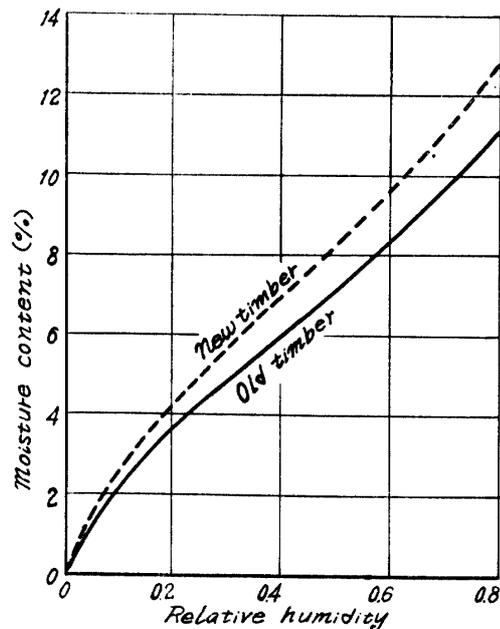


B Heat treated wood (HINOKI, tangential, 100°C, oven dry)

Fig. 1 Shrinkage



A Old timber (HINOKI)



B Heat treated wood (HINOKI, 130°C, Oven dry, 5hr.)

Fig. 2 Absorption isotherm

averaged through these repetitions. As the basis of the percentage, of shrinkage, the tangential length at the first drying has been taken.

2-2. Hygroscopicity

Fig. 2 A shows an example of the comparative absorption-isotherms of water vapour on the old timber and on the new timber. The old timbers showed a poorer hygroscopicity than the new ones. The differences of hygroscopicities

within the old and within the new were negligibly small.

The procedures of the investigation are as follows. In an evacuated tube, the specimens sized 1.8×8×60mm with radial surface were set on quartz-fibre springs. The measured amounts of the vapour were sent step by step into the tube, and the equilibrium was set up at every intrusion. The absorption isotherm was

obtained from the equilibrium weight of the specimen and the pressure in the tube.

2-3. Mechanical properties ^{1) 7) 8) 10) 13) 15)}

Some of the mechanical properties of the

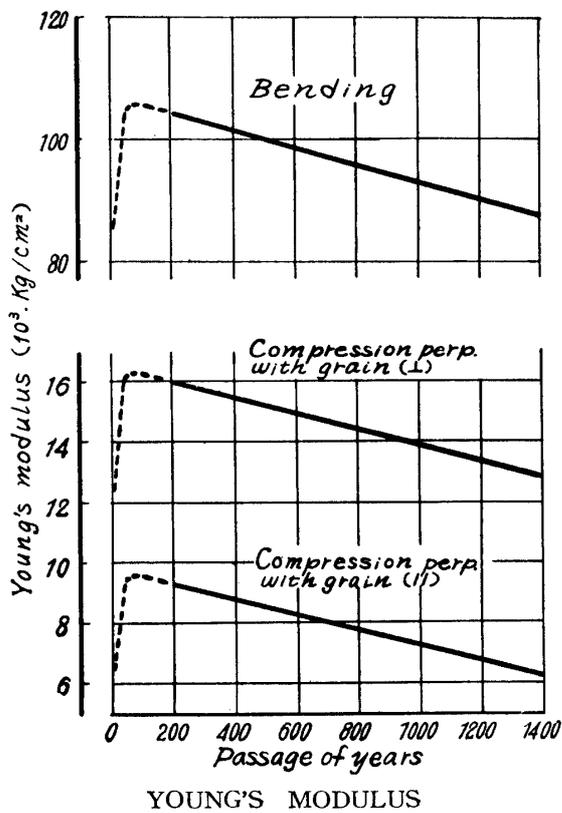
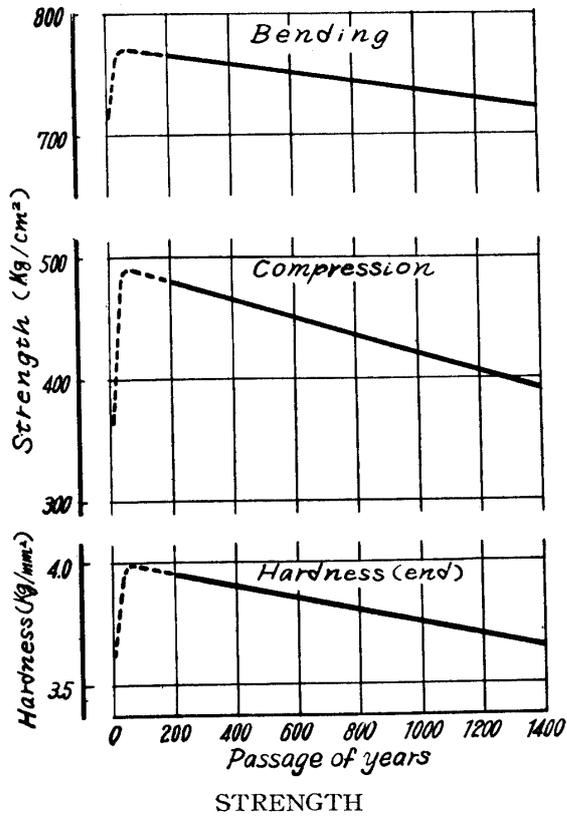


Fig. 3 A Mechanical properties, SOFT WOOD (HINOKI)

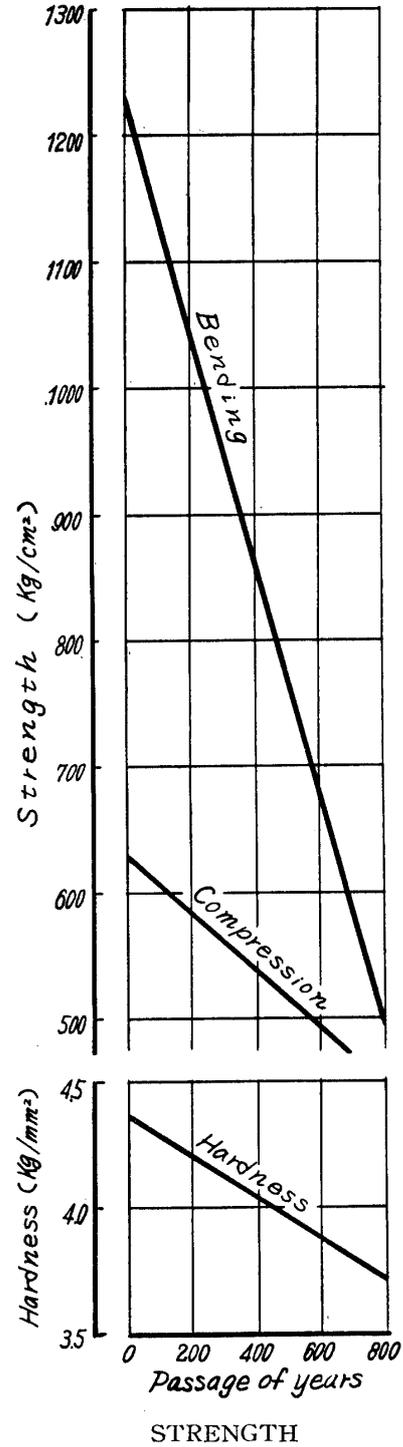
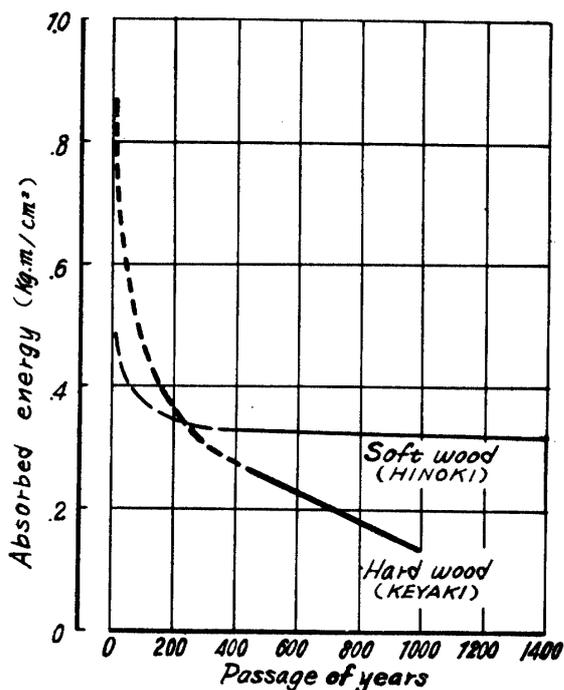


Fig. 3 B Mechanical properties, HARD WOOD (KEYAKI)

old timbers of Hinoki, such as compression, hardness, and Young's modula, seemed stronger than those of the new timber. After the corrections of strength values for moisture content and specific gravity, the series of the corre-



STRENGTH (impact bending)

Fig. 3 C Mechanical properties (HINOKI & KEYAKI)

sponding strength-age values were plotted with respect to vertical and horizontal axes, then it was found that the average curves showed maxima, as given in Fig. 3 A.

Fig. 3 B gives some of the corresponding strength values of the old timbers of Keyaki. In this figure, the values are not corrected but the trend of decrease is obvious.

Impact bending, cleavability, and shear reduced evidently with time, even in the timbers of Hinoki. The reduction was still more vigorous in Keyaki. For example, Fig. 3 C shows the reduction of the impact bending of those two kinds of timber with their age.

2-4. Analysis of constituents ^{5) 11)}

The trend of the content of each constituent of the timbers with their age is given in Fig. 4. In both of these two kinds of wood, the soluble constituents increase with time, and the contents of the cellulosic materials decrease. The apparent constancy of the pentosan content might be due to the procedure of its estimation in terms of furfural.

The rate of increase or decrease of each constituent in the timber of Hinoki, is com-

pared with the corresponding rate in Keyaki, in Fig. 5 A. On an average, those rates in Keyaki are about three times greater than the corresponding rates in Hinoki. The rate of decrease of the cellulosic materials in the former is nearly five times greater than the rate in the latter. These differences obviously correlate with those of the reduction rates of the mechanical properties between two species of wood.

2-5. Analysis of unearthed wood ^{14) 16)}

The analysis of a soft wood and a hard wood, both of which were unearthed from the same spot, indicates the difference between the kinds of wood, destroyed under the same condition.

Fig. 6 shows the two examples of such couples: Fig. 6 A is on the timbers about two thousand years ago, unearthed from KARAKO Sites in NARA Prefecture, and Fig. 6 B is on the wood specimens from the TUNDRA Districts in SOUTH SAGHALIEN, which are estimated to be aged tens of thousands of years.

With these two couples and others, the cellulosic materials in hard woods were found to have been much more destroyed than those in soft woods.

2-6. Colour ²⁾

The pale cream-yellow of the timber of Hinoki takes a brown tint with time, uniformly in its inner part under the surface layer several millimetres thick.

The transparency of the alcohol-extract solution of the timber shows a reduction with its age. Fig. 7 A gives the extinction of the alcohol-extract solution, increasing almost linearly with time. Accordingly, the amount of the extracted colouring material is estimated to increase in proportion to the other soluble constituents as above.

The luminosity of the tangential surface, freshly prepared from the timber is given in Fig. 7 B. The luminosity was determined with trichromatic colorimeter based upon the CIE system. The trend of the luminosity is in accordance with the increasing extinction or, conversely, with the reduction of the transparency of the alcohol-extract solution.

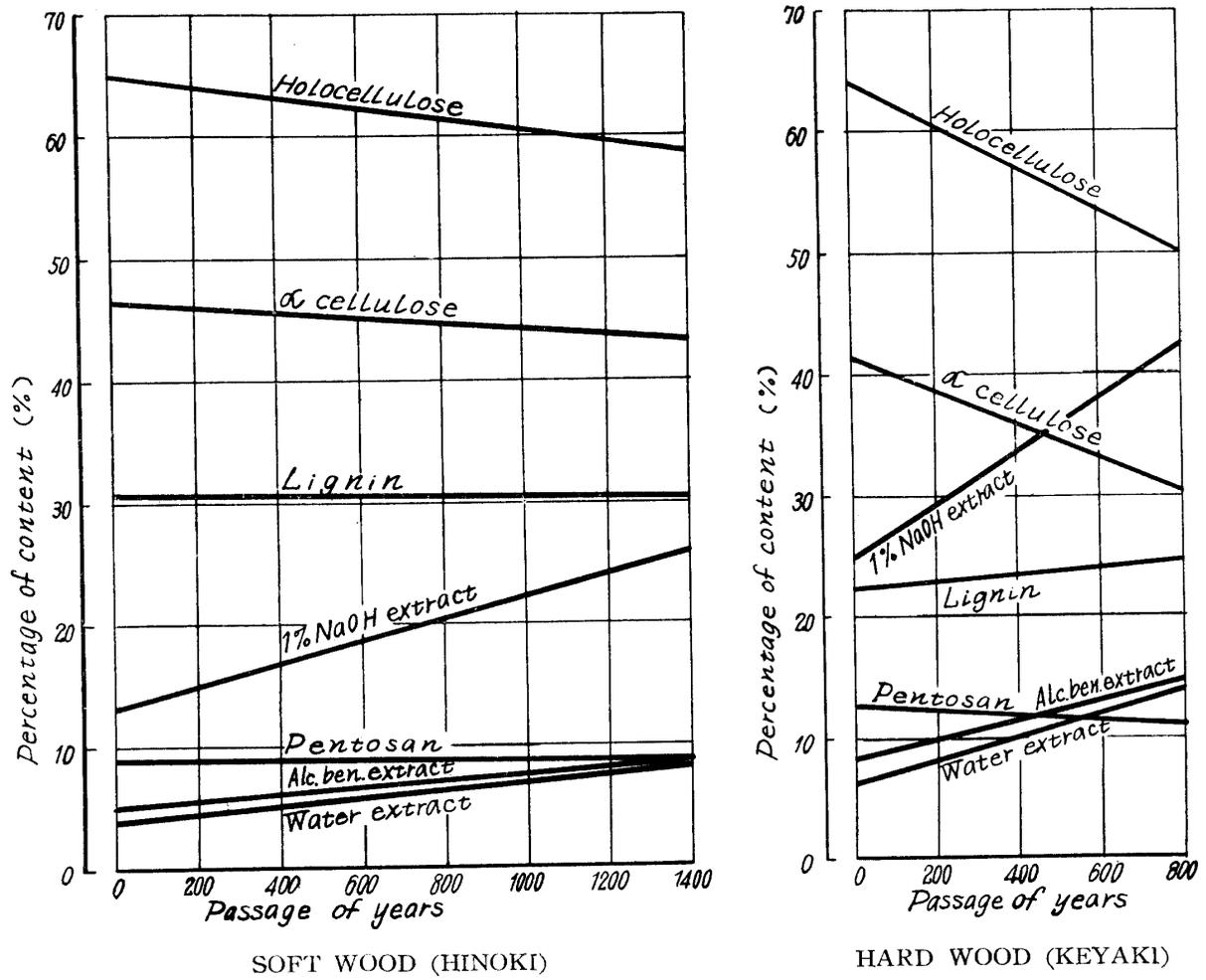


Fig. 4 Analysis of old timbers

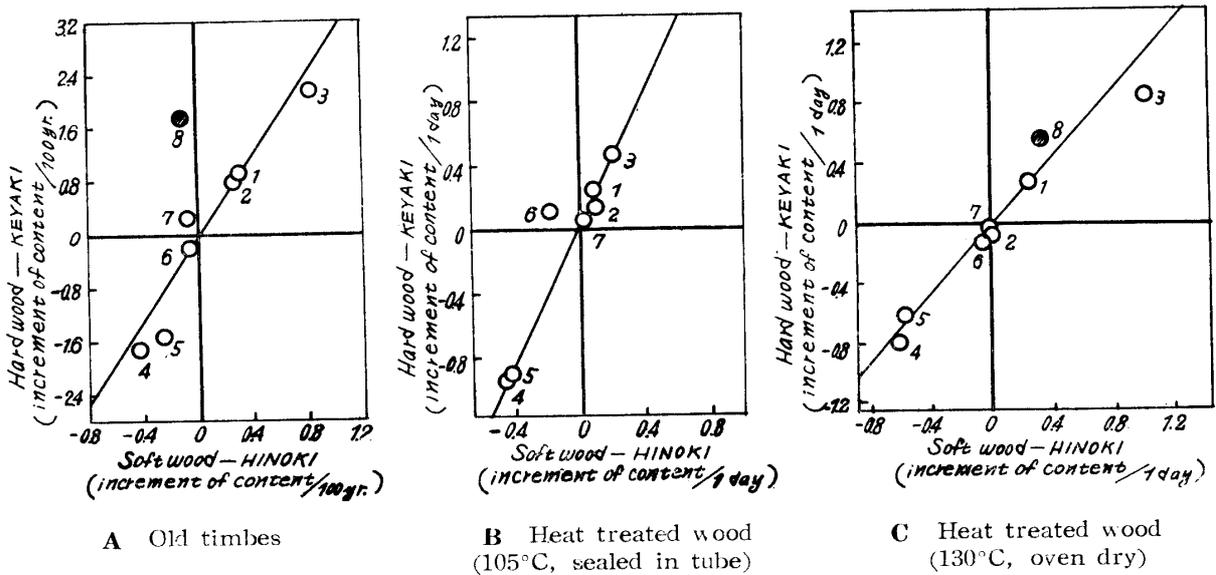


Fig. 5 Increase or decrease of constituents of soft wood and hard wood
 1. Water extract, 2. alcohol benzen extract, 3. 1%NaOH extract;
 4. holocellulose, 5. α -cellulose; 6. pentosan, 7. lignin; 8. reduction of weight (original sample basis; A, estimated from specific gravity)

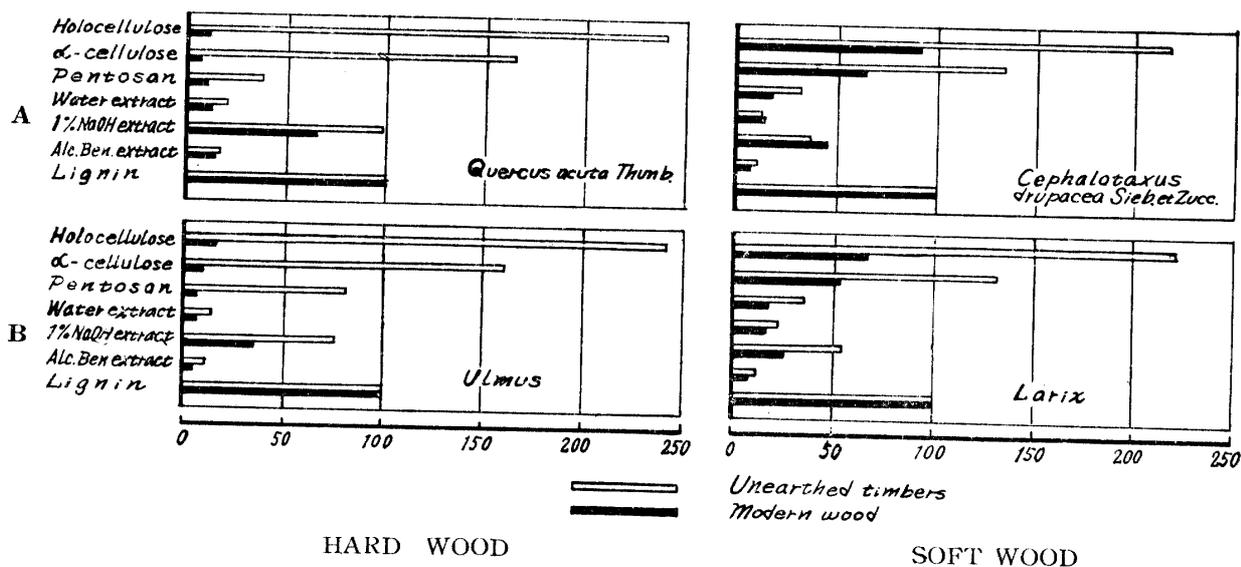
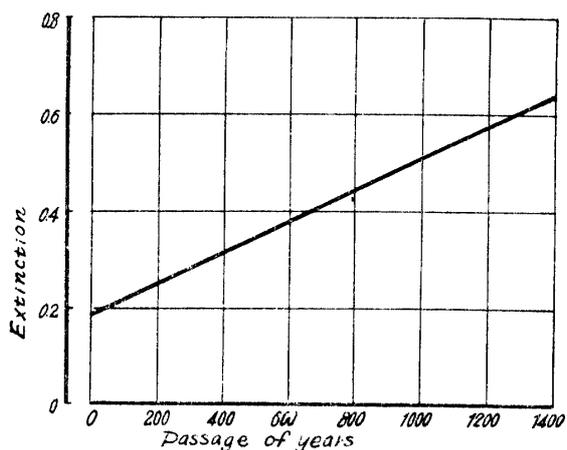


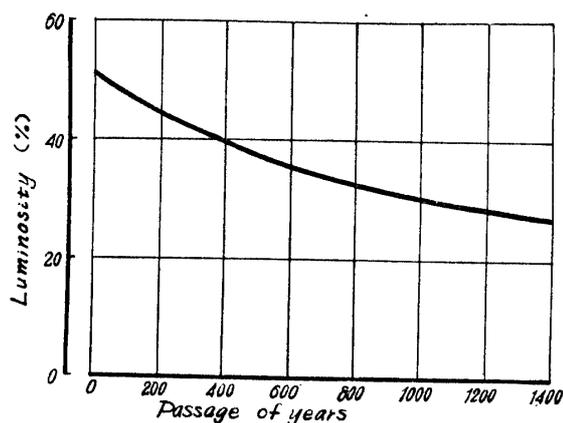
Fig. 6 Analysis of timbers and woods buried under earth (lignin basis)

A : Timbers unearthed from the KARAKO sites

B : Woods from tundra districts in Saghalien



A Extinction of alcohol extract solution



B Luminosity of tangential surface

Fig. 7 Colorimetry (HINOKI)

The chromaticity of the timber showed a slight transition from green to red with its age.

2-7. Content of the crystalline-region of cellulose ³⁾

The cellulosic materials decreased their content in the timber with time, as was mentioned above. There, by the usual method of analysis, the qualitative difference between the new timber of Hinoki and the old one, such as α -cellulose content based on the original holocellulose, was not detected. But the investigation of their accessibility, following the method of BATTISTA (1950), and that of their viscosity in the cuprammonium indicated the qualitative difference be-

tween the new and the old.

As a measure of the chemical inertness or, inversely, the accessibility, BATTISTA has taken the amount of the residue, obtained from the cellulosic material, which has been boiled in hydrochloric acid. The yield of the residue may be named the "content of crystalline region of cellulose", or briefly the "crystalline-cellulose content".

The crystalline-cellulose content based on the original holocellulose is higher in the old timbers, than in the new ones, and the differences of the content within the old and within the new were not detected. These results

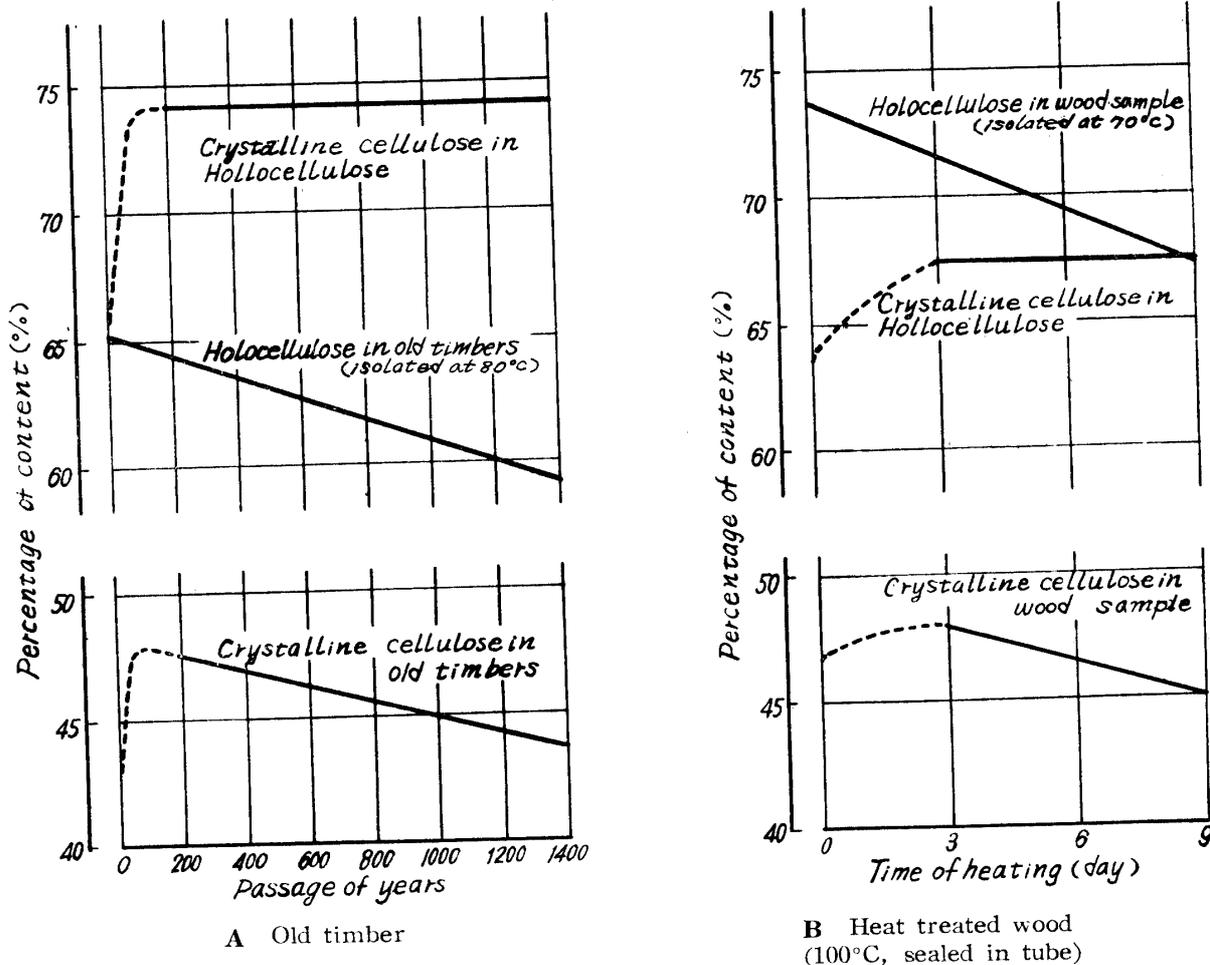


Fig. 8 Crystalline cellulose content (HINOKI)

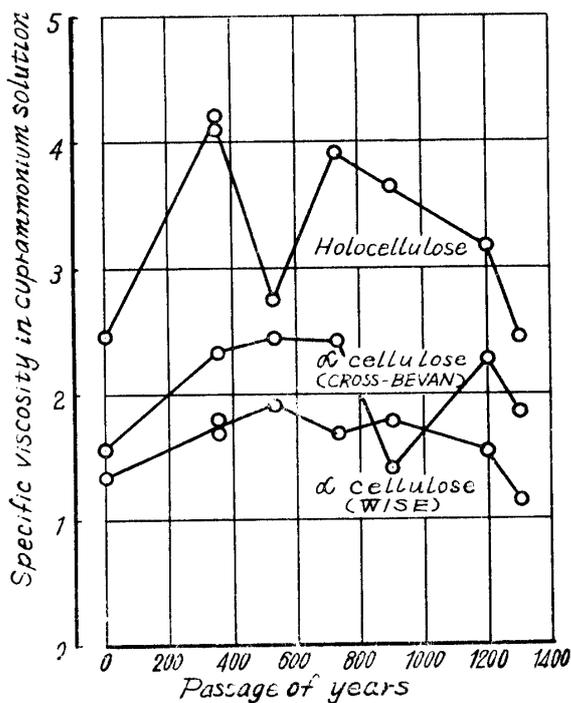


Fig. 9 Viscosity of cellulose solutions (HINOKI)

are shown in Fig. 8A together with the content based on dry wood. The variation in the crystalline-cellulose content based on dry wood has a significant correlation with the strength of the timbers given in Fig. 3A.

2-8. Viscosity of the cuprammonium solution of cellulose⁽⁴⁾

Fig. 9 gives the viscosities of the cellulosic materials of the timber of Hinoki. The concentration of the solution was 0.05 grams of the air-dried sample in 10cc of the solvent. As the samples of the cellulosic materials, holo- and α -cellulose of WISE and α -cellulose of CROSS-BEVAN were taken.

Through these three modifications, the specific viscosity of the materials increase and then decrease with the age of the timber. The trend is analogous to that of the crystalline-cellulose content based on dry wood, and that of the mechanical strength.

III. DISCUSSION

3-1. Old timber and heat-treated wood

With almost all the data above cited, the corresponding data on the heat treatment have been obtained, and they are given in Figures 1B, 2B, 5B, 5C, 8B and Fig.10. The qualitative resemblance between the processes under these two groups of condition are obvious.

The crude comparisons of the available data of the rates in the old timber with the corresponding rates in the heat-treated wood are tried in Table 2. Through the degradation of the cellulosic materials and some other changes, the rates are magnified up to $10^4 \sim 10^5$ times of those in the old timber, by the heating. These ratios correspond to about 25 kcal/mole of the energy of activation, if ARRHENIUS's formula on the rate of chemical reaction might be adopted. According to this estimation of the effect of temperature on the rate, the rate under saturated vapour would be nearly ten times of the rate under oven-drying.

As shown in Figures 5B and 5C, the rate of increase in the content of the soluble constituent on heating stood at somewhat lower proportion to the rate in the old timber, than the corresponding proportion found in the cellulosic materials. This apparent discrepancy may be due to the evaporation of the constituent, and the resemblance between the old timber and the heat-treated wood is expected to be more per-

fect.

These considerations on the resemblance suggest that the variation in quantity of the cellulosic materials with time might be one of the main factors of other changes.

3-2. Mechanical properties and crystalline-cellulose content

The corrected values of the strength properties, given in Fig. 3A, were obtained by the following calculations. The strength value, U , observed on the specimen, was tried to be fitted on the regression equation,

$$U = a(x-45) - b(y-13) + c(z-40) + u,$$

where x is the crystalline-cellulose content based on dry wood, y is the moisture content at the testing time, z is one hundred times of the specific gravity based on dry wood, and a , b , c , and u are the parameters determined by the method of least squares. Then, the corrected value is equal to

$$U + b(y-13) - c(z-40).$$

The abscissa of the average curve is equal to

$$u + a(x-45).$$

The coefficients, b and c , which represent the effect of the moisture on the strength and that of the specific gravity respectively, were coincident with the data on the new timbers obtained by many workers. The deviations of the corrected values of compression and Young's modulus of bending from the average line were found to be of the same order of magnitude as the devia-

Table 2. The relative rate of change in the heat-treated wood based on the corresponding rate in the old timber

HINOKI			
Relative rate	Temperature	Humidity	Source of Estimation (Reduction rate of)
6.3×10^4	130°C	Oven dry	Holo- and α -cellulose contents ¹²⁾
5.5	130	Oven dry	Young's modulus of bending ⁶⁾
1.3	100	Oven dry	Shrinkage
4.7	105	Sealed in tube	Holo- and α -cellulose contents
7.5	100	Sealed in tube	Holocellulose content ³⁾
KEYAKI			
1.2	130	Oven dry	Holo- and α -cellulose contents ¹²⁾
2.1	105	Sealed in tube	Holo- and α -cellulose contents

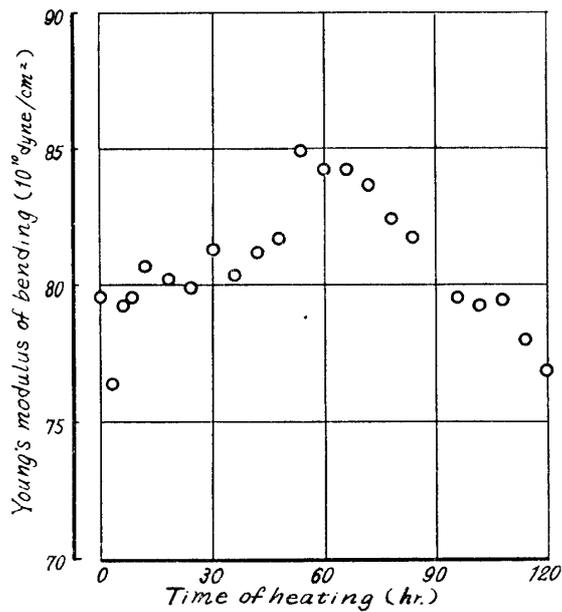


Fig. 10 Young's modulus of bending of heat treated wood (HINOKI, 130°C, oven dry)

tions within the new timbers, which are estimated from the data of MIYOSHI (1951).

The statistical significance of the regression coefficient, a , indicates that the corrected value of the strength first increases and then decreases with the age of the timber, in proportion to the variation in the crystalline-cellulose content based on wood. Accordingly, it is evident that the strength increases with the chemical inertness of the cellulosic materials at the beginning of

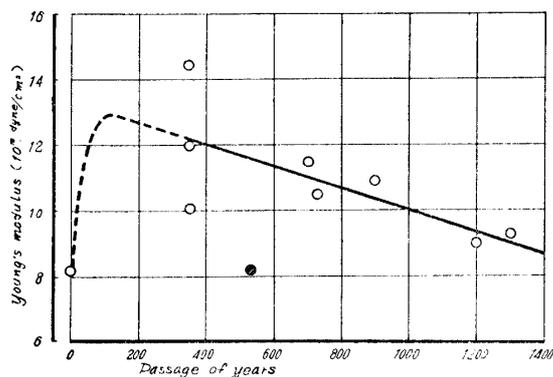


Fig. 12 Dynamic Young's modulus of bending of old timbers (HINOKI)

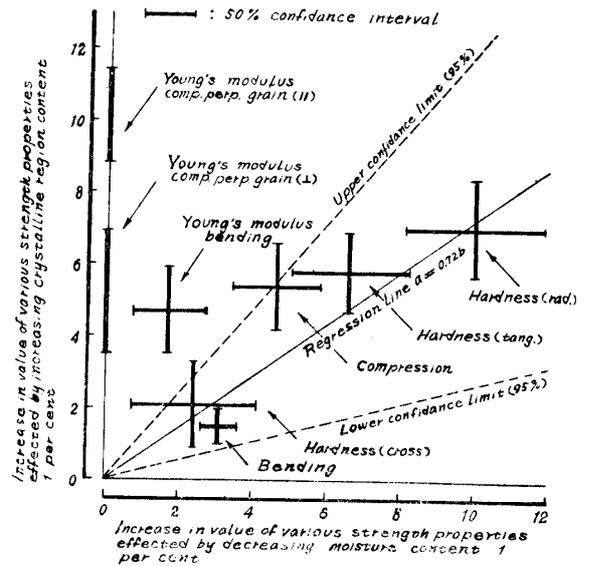


Fig. 11 Increase (or decrease) in values of various strength properties effected by moisture content and by crystalline region content. *

the ageing, and, after the saturation of "crystallization", it decreases with the cellulose as a whole. This conclusion may be accepted, however the observed "crystalline-cellulose content" might be interpreted.

If the observed "crystalline-cellulose content" is taken to be nearly right measure of the crystalline region in a submicroscopic picture of the cellulose, the following three considerations, besides the above mentioned observation, are

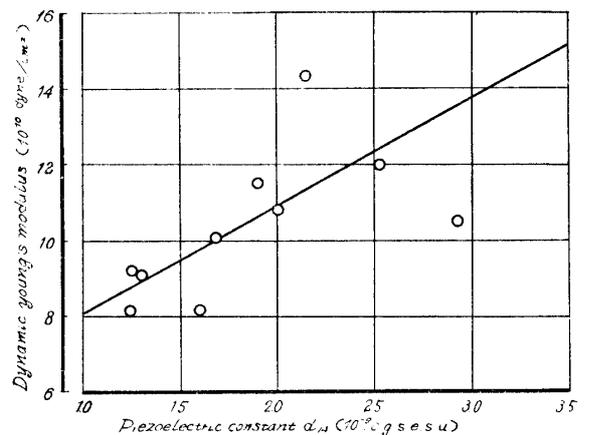


Fig. 13 Dynamic Young's modulus of bending and piezoelectric constant of old timbers (HINOKI)

*The coefficients a and b have been roughly estimated without any consideration about error of x, y, z , and U . Their reexamination may be reported presently.



Fig. 14 The Hōdō in the Byōdōin Temple

consistent with this view.

(1) The reduction of the hygroscopicity in the old timber, as given in Fig. 2A, might be due to the decrease in the amount of the amorphous region or, inversely, the increase in the crystalline region.

(2) The coefficient, a , in the regression equation has a trend of increase with the coefficient, b , for the destructive strength, as shown in Fig. 11. That is, the submicroscopic change in the structure of the cellulose and the mechanism with which the submicroscopic structure affects the strength of wood might be common to both cases, the decreasing moisture-content and the increasing "crystalline-cellulose content." For Young's modulus, the trend has been an expected one, but the ratio a/b seems to be much larger than that of the destructive strength.

(3) The rate of increase in the "crystalline-cellulose content" and the rates in such strength values as above cited, at the beginning of the

ageing, were estimated only on the poor basis because of the lack of the specimens not so old. From the comparison between new timbers, each of which was cut down a few years sooner or later than others, the rate of 0.5~1.0 per cent increment of "crystalline-cellulose content" a year was obtained. With respect to the rate of increase in Young's modulus in Fig. 10 and the rate of increase in the crystalline-cellulose content in Fig. 8B, this value gives the energy of activation of about 15 kcal/mole. The energy value obtained seems reasonable, when the process is supposed to be due to the rearrangement of cellulose molecules.

The results of the investigation of the viscosity, might suggest another possible scheme, in which the degrees of polymerization, estimated from the viscosity, are taken as a variable. At present, the crystalline-cellulose content is preferred in place of the degrees of polymerization, as the observed value of the viscosity might be

affected by the accessibility of the cellulosic materials as well as by their intrinsic degrees of polymerization.

Recently, one more basis consistent with this view has been obtained by FUKADA. He observed the dynamic Young's modulus and piezoelectric constant of our old timbers. The latter also shows such variation with time as the former. The results are given in Figures. 12 and 13.

3-3. Soft woods and hard woods in their natural durability

It has been commonly known with the repairs of old edifices in Japan, that the timber of Hinoki endures longer than that of Keyaki.

One of the examples may be drawn from the recent renovation of the Hoo-do, a building symbolising a phoenix shown in Fig. 14, in the Byodo-in Temple near Kyoto. All of the four beams, which run horizontally along the diagonal lines of the main portion of the building and hold the weight of the eaves projecting widely over, have been found to be broken down right in two. They are almost all of the few members of Keyaki, in the predominance of Hinoki. It is believed that the kinds of wood were selected in accordance with the load and strength of the members, but without any precaution about their permanence, at the time of the erection about nine hundred years ago.

In this report, it has been established that the content of the cellulosic materials in Keyaki decreases faster than that in Hinoki, and the initially superior strength of the former drops down below that of the latter within several centuries, under the conditions free from biological agency. It has also been shown by the analysis of the unearthed woods, that the content

of the cellulosic constituents in the hard woods is much less than that in the soft woods. The difference of the rate of decomposition of the cellulosic materials between these two species has also been produced under the artificial condition, with the same temperature and humidity, as in Fig. 5 B.

Now, it is known in studies on the microstructure of coal that the cell texture remains in soft woods, while it is not detected in hard woods. It is also widely known in pulp industry that hard woods are treated by reagents with less difficulty than soft woods.

Then, the difference of permanence, between the timber of Hinoki and that of Keyaki, is expected to represent the general difference between soft woods and hard woods.

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