Title	<Note>Some Physical Properties of Wood and Cellulose Irradiated with Gamma Rays
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Citation	Wood research : bulletin of the Wood Research Institute Kyoto University (1977), 62: 19-28
Issue Date	1977-02-28
URL	http://hdl.handle.net/2433/53373
Right	
Type	Departmental Bulletin Paper
Textversion	publisher

NOTE

Some Physical Properties of Wood and Cellulose Irradiated with Gamma Rays

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Abstract——The effect of gamma irradiation from cobalt-60 on specific gravity, degree of crystallinity, thermal softening temperature, tensile strength and torsional creep behavior of wood and cellulose were investigated and the following results were obtained.

1) A dosage up to about 10^8 rad had little effect on the specific gravity of wood and cellulose.

2) The degree of crystallinity of wood and cellulose remained almost unchanged up to 3×10^7 rad, but started to decrease rapidly at about 1×10^8 rad.

3) The softening temperature of cellulose shifted gradually to a lower temperature region up to about 3×10^7 rad and abruptly in the range exceeding 3×10^7 rad.

4) Strength of wood decreased with increasing irradiation dosage, depending remarkably on loading modes.

5) The value of creep compliance of wood at time 0.1 min did not change up to 1×10^7 rad, but increased markedly in the range exceeding about 3×10^7 rad.

Introduction

High energy radiation causes changes in the chemical[1~16] and crystalline structures of wood, but does not result in significant changes of its anatomical or supramolecular structures[38~48], when radiation dosages up to 5×10^8 rad are used. As the physical and mechanical properties[17~37] of wood depend on its chemical and crystalline structures, some attempts applying high energy radiation techniques to modify and improve the hygroscopicity, swelling, and decay susceptibility of wood have been made. However, prior to this an understanding of the effects of high energy radiation on the physical and mechanical properties in relation to the chemical structure is necessary. We have investigated the relationship between dielectric properties and the molecular structure of wood irradiated with gamma rays[56~58]. In this paper, we outline the changes in some physical properties of wood and cellulose induced by the irradiation of gamma rays in the light of the many published references[1~58].

Experimental

The samples used were Hinoki wood (*Chamaecyparis obtusa* ENDL., specific

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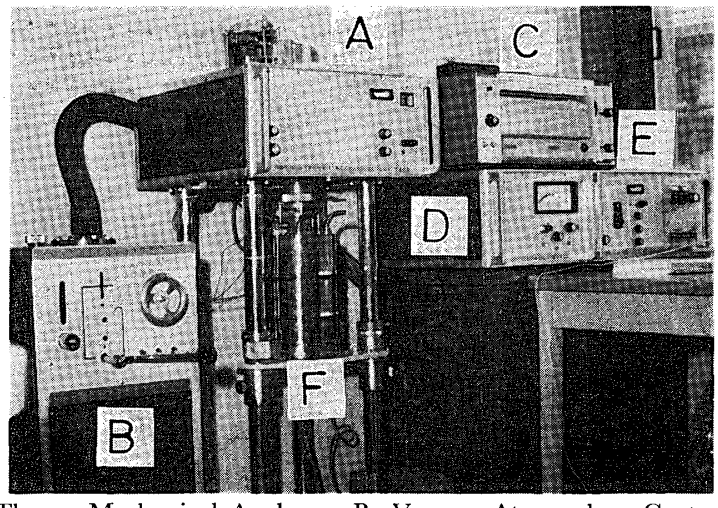
gravity : 0.41), Hoonoki wood (*Magnolia obovata* THUNB., specific gravity : 0.54) and Whatman cellulose powder irradiated with gamma rays from cobalt-60 under an air atomosphere for 0, 1, 3, 10, 30, 100 hours at a dose rate of 3.12 Mrad/hr for wood and at 3.20 Mrad/hr for cellulose.

The creep measurements were carried out at 20° and 50°C in water using a torsional apparatus[59]. The dimension of the specimen was 6.60 (L) × 1.00 (R) × 0.110 (T) cm. The tensile strength of the irradiated wood was measured in water at 20°C using an instron type tensile testing instrument (TOM 5000X produced by Shinko Ltd.). Furthermore, the amount of thermal expansion for the irradiated cellulose under a constant compressive load (10 g) was measured in vacuo in the temperature range of 20° to 300°C at a heating rate of 2°C/min using a thermo-mechanical analyzer (TM 1500, Sinku Riko Co., Ltd., see Fig. 1).



A: Thermo-Mechanical Analyzer, B: Vacuum-Atomosphere Controller, C: Recorder, D: Stress Relaxation Circuit, E: Thermal Program Controller, F: Temperature Controlled Chamber.

Fig. 1. Photograph of Thermo-Mechanical Analyzer TM 1500.

On the other hand, in order to make clear the relationship between crystalline structure and mechanical properties of the irradiated wood and cellulose, X-ray diffraction measurements were performed by the transmission method (Cu-Kα) using a Rotaflex (RU-3L X-ray diffractmeter, Rigaku Denki Co., Ltd.), and the relative degree of crystallinity was estimated by the crystallinity index defined by Segal and others[60].

Results and Discussion

1) *Specific Gravity*

In Fig. 2, the relative specific gravity of the irradiated wood ρ_i/ρ_0 is plotted against logarithumic irradiation dosage log D. In this figure, the data of woods

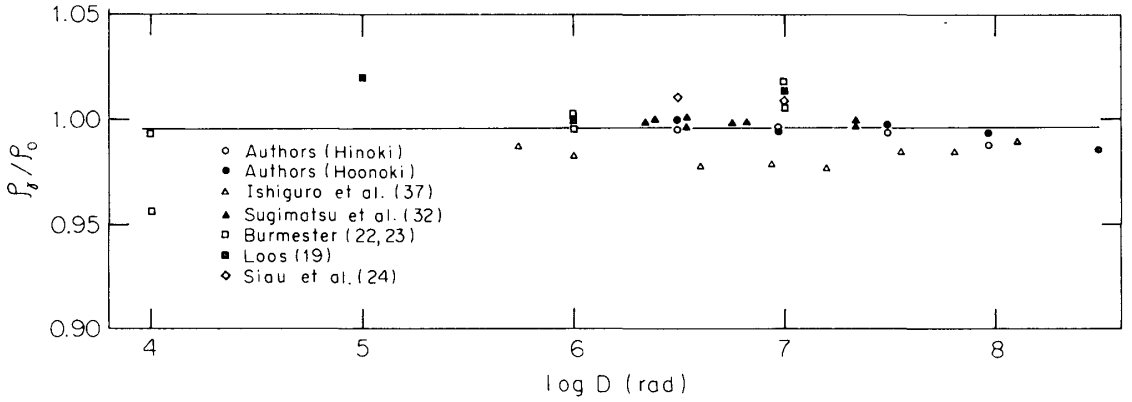


Fig. 2. Relative specific gravity ρ_r/ρ_0 of wood and cellulose as a function of irradiation dosage log D. ρ_r: irradiated, ρ_0: untreated.

and cellulose published by many authors[19,22,23,24,32,37] are also included. SUGIMATSU *et al.*[32] reported that the value of specific gravity of cellulose slightly increases at about 4×10^6 rad when cellulose is irradiated with gamma rays under an oxygen atomosphere. On the other hand, the data published by ISHIGURO *et al.*[37] showed that the value of specific gravity of wood irradiated with gamma rays in vacuo decreases once up to 1×10^7 rad and then increases at irradiation levels in excees of 1×10^7 rad. However, as is evident from the figure, it seems that a dosage up to about 10^8 rad has little effect on the specific gravity of wood and cellulose.

2) *Degree of Crystallinity*

TAKAMUKU *et al.*[8] reported that the dosage of crystallinity of cellulose is almost unchanged up to 5×10^7 rad. MURAYAMA[34] showed that in X-ray diffraction measurement any changes in the lattice constant of cellulose can not be detected at a dosage of 1×10^8 rad. HIRAI *et al.*[35] also reported that the degree of crystallinity of wood remains unchanged up to 1.4×10^8 rad. However, the result published by SEIFERT[14] showed that the degree of crystallinity of wood slightly decreases at a dosage of $10^{8.3}$ rad in comparison with that of the untreated wood. Furthermore, ISHIGURO *et al.*[37] showed that the degree of crystallinity starts to decrease rapidly at about 1×10^8 rad, and GOTO *et al.*[48] comfirmed that the crystalline peaks observed in the untreated wood disappear completely at a dosage of 6.55×10^8 rad.

In Fig. 3, thc relative crystallinity index β_r/β_0 of wood and cellulose as a function of log **D** are shown. The data published by many authors[8,34,35,37,48] are also plotted in this figure. β_r/β_0 value of wood and cellulose almost remains unchanged up to 3×10^7 rad, but starts to decrease rapidly at about 1×10^8 rad. And, the value falls to 0.1 in cellulose and 0.5 in wood at about 1×10^8 rad. A similar tendency can be detected in the infrared spectra in which a broadening of the so-called crystalline sensitive bands (1110, 1060, 1040 cm^{-1})[61] occurs at 9.7×10^7 rad. From

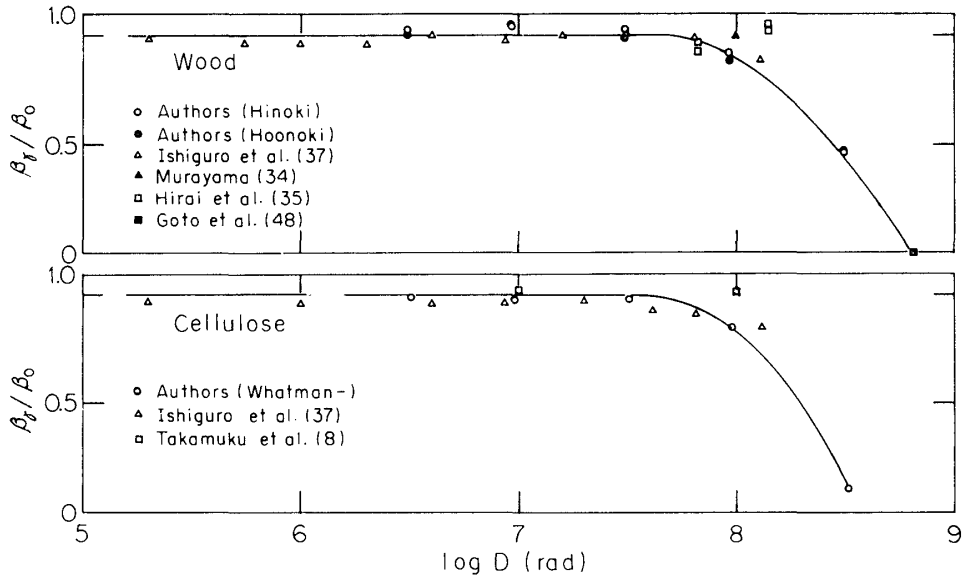


Fig. 3. Relative crystallinity index β_γ/β_0 of wood and cellulose as a function of log D.

the figure, it seems that the crystallinity of wood decreases gently compared with that of cellulose in the range exceed 10^8 rad. Consequently, it may be concluded that wood is more resistant to degradation due to high energy radiation than cellulose because of the presence of encrusting substances such as lignin and extractives.

3) *Degree of Polymerization*

SAEMAN *et al.*[1] studied the effect of high energy cathode ray irradiation on wood, wood pulp and cotton linters, and showed that the effect of irradiation on degree of polymerization **DP** for cellulose is not appreciable up to a dosage of 10^6 equivalent roentgens, but at high dosage the rapid decrease in **DP** occurs. CHERLESBY[3] used the viscosity data of SAEMAN *et al.*[1] to verify a theoretical relation between intrinsic viscosity and radiation dosage. MIZUKAMI[9] and HORIO *et al.*[13] found that the plots of logarithum of the difference between the reciprocal of **DP** of the irradiation cellulose and that of the untreated cellulose against log **D** ranging from 5.2×10^5 to 1.1×10^8 roentgens give a straight line. On the other hand, HUANG *et al.*[10] reported that an approximately linear relation is obtained when **DP** is plotted against gamma irradiation dosage on a log-log scale, and this result was in good agreement with that of BLOUIN and others[6]. Furthermore, NEAL *et al.*[12] showed that the number of bond broken by electron irradiation is a linear function of irradiation dosage for wood pulp.

In Fig. 4, **DP** value published by many authors[1,6,8,9,10,12,13,21,32] is plotted against irradiation dosage on a log-log scale. From this figure, it is evident that there is a linear relationship between log **DP** and log **D** in the region from 10^6 to 10^8 rad.

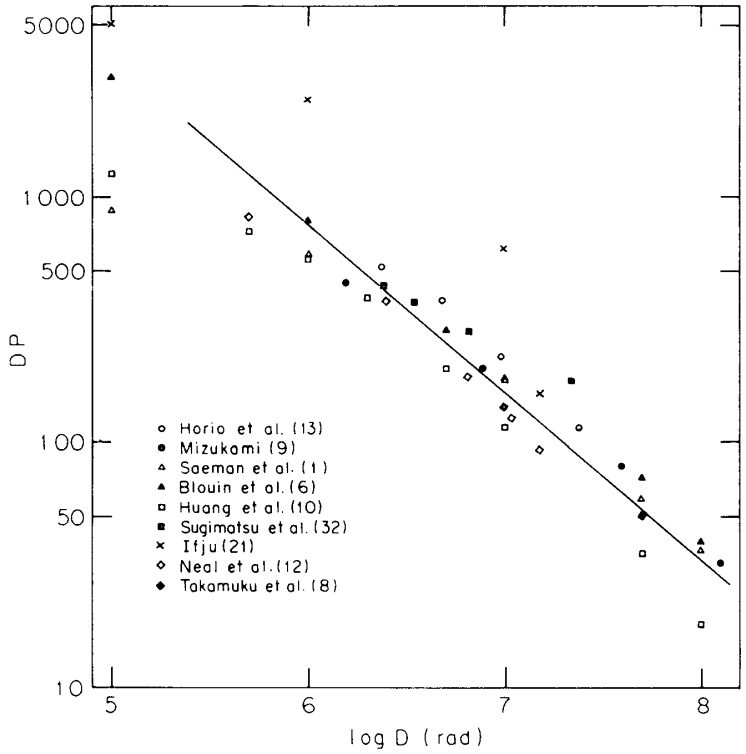


Fig. 4. Degree of polymerization DP of cellulose as a function of log D.

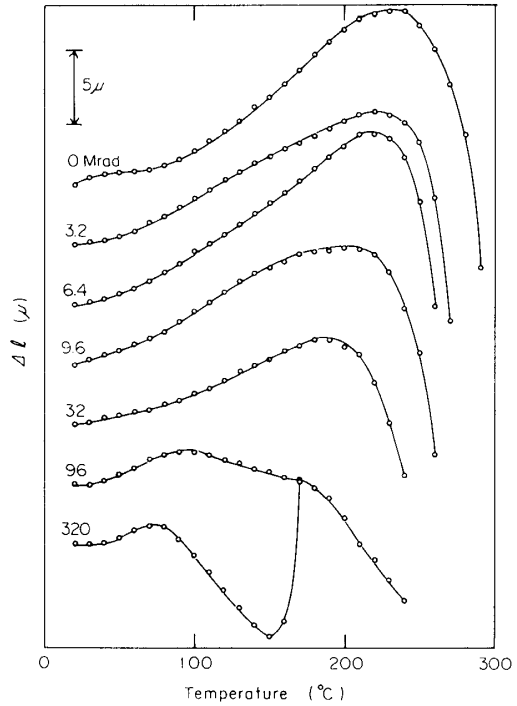


Fig. 5. Thermal expansion of gamma irradiation Whatman cellulose under a constant load.

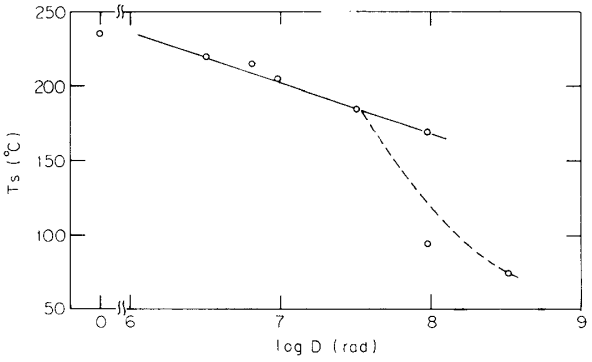


Fig. 6. Softening temperature Ts of Whatman cellulose as a function of log D.

4) *Thermal Softening Temperature*

Fig. 5 illustrates the amount of thermal expansion under a constant compressive load for Whatman cellulose irradiated with gamma rays. When the amount of expansion was plotted against temperature, a clear maximum occurred. The peak position on the temperature scale, which is defined by GORING[62] as a softening temperature **Ts**, was about 235°C for untreated cellulose, which is in good agreement with that of GORING[63]. Fig. 6 shows **Ts**-log **D** curve of Whatman cellulose. **Ts** decreases linearly up to about 3×10^7 rad, where any changes in the degree of crystallinity can not be detected, but **DP** value decreases linearly against log **D**. Therefore, it is assumed that **Ts**-shift to lower temperature range would be due to the scission of chemical bond such as a glucosidic bond. On the other hand, **Ts**-shift in the range exceeding 3×10^7 rad would be due to degradation of crystalline structure.

5) *Strength*

SIAU *et al.*[24] reported that there is no apparent reduction in compressive strength of wood perpendicular to grain up to 1×10^7 rad and KARPOV *et al.*[18] showed that degradation starts at 1×10^7 rad in bending and 1×10^8 rad in compression. Furthermore, BURMESTER[22,23] found that there is an insignificant increase in tensile and compressive strengths resulting from develop of hydrogen bond in wood when low gamma irradiation dosages are used, but with increasing dosage tensile strength reduces more rapidly than compressive strength. RAMALINGAM *et al.*[20] also found that the percent loss in bending strength of wood is directly propotional to gamma irradiation dosage beyond the level of 2×10^6 rad. SEIFERT[14] reported that the changes in the chemical properties reflect strength of the gamma irradiated wood and the decrease in tensile and compressive strengths is due to the reduction of NaOH insoluble fraction of wood up to about 10^8 rad. On the other hand, IFJU[21] found that tensile strength of gamma irradiation wood depends mostly on cellulose **DP** and the decrease in **DP** reduce strength more in low **DP** region than in high **DP** region.

In Fig. 7, relative compressive strength $\sigma_{\gamma c}/\sigma_{0c}$, bending strength $\sigma_{\gamma b}/\sigma_{0b}$ and tensile strength $\sigma_{\gamma t}/\sigma_{0t}$ of wood are plotted against log **D** with the results reported by other authors[14,18,19,20~24]. The results reported by GILFILLAND *et al.*[17] showed that the removal of water and oxygen by evacuation dose not have any effect on the strength of the beta irradiated cotton yarns. Furthermore, the study of IFJU[21] on moisture content and temperature dependence of tensile strength of gamma radiation wood showed that tensile strength of wood with degraded cellulose is more sensitive to changes in moisture content. SIAU *et al.*[54] also emphasized that the results published by KARPOV *et al.*[18], RAMALINGAM *et al.*[20] and KRANOVITSKAYA,

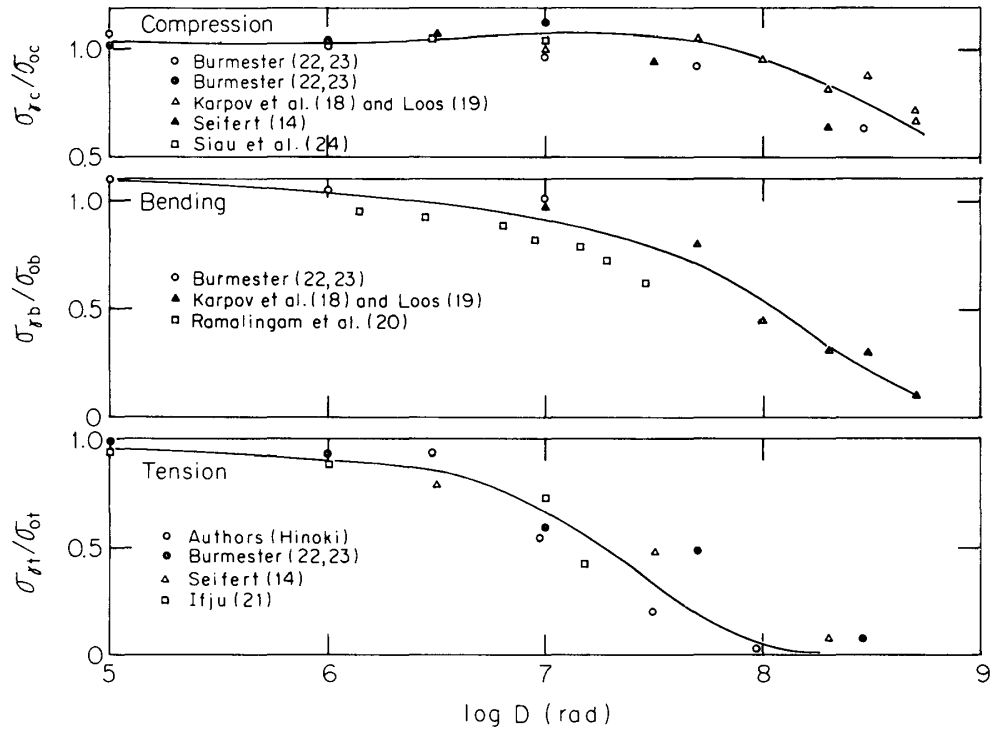


Fig. 7. Relative compressive strength $\sigma_{\gamma c}/\sigma_{0c}$, bending strength $\sigma_{\gamma b}/\sigma_{0b}$ and tensile strength $\sigma_{\gamma t}/\sigma_{0t}$ of wood as a function of log D.

differ widely and this results from different measuring conditions. However, from Fig. 7, it is evident that both temperature and moisture content have little effect on strengths, but strength depends remarkably on loading modes. In this connection, the relative strength decrease in the order, tensile strength>bending strength> compressive strength.

6) *Viscoelastic Properties*

The results reported by SUGIMATSU *et al.*[32] shows that the value of dynamic elastic modulus reached a maximum at around 5×10^6 rad when cellulose was irradiated under an oxygen atomosphere, and they atribute it to the formation of inter-cellular bonding. On the other hand, SULER[36] showed that the value of dynamic bending elastic modulus of wood remains unchanged up to 1×10^7 rad, and ISHIGURO *et al.*[37] also showed that the ratio of dynamic elastic modulus to specific gravity as well as the degree of crystallinity rapidly starts to decreases at about 3×10^7 rad. In addition CHOONG *et al.* reported that the value of bending elastic modulus is almost unaffected by gamma irradiation[55]. On the contrary, HIRAI *et al.*[35] showed that the value of dynamic compliance of wood increases by about 90 percent at 2.5×10^8 rad.

Fig. 8 illustrates the torsional creep curves of gamma irradiation Hinoki wood in water at 50°C. Furthermore, Fig. 9 shows the values of creep compliance $\mathbf{J}(0.1)$

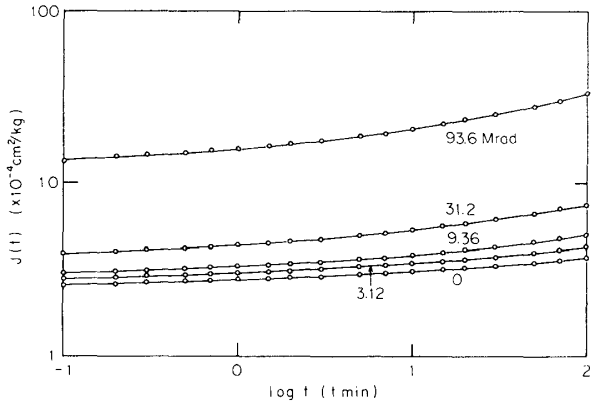


Fig. 8. creep curves of gamma irradiation Hinoki at 50°C in water.

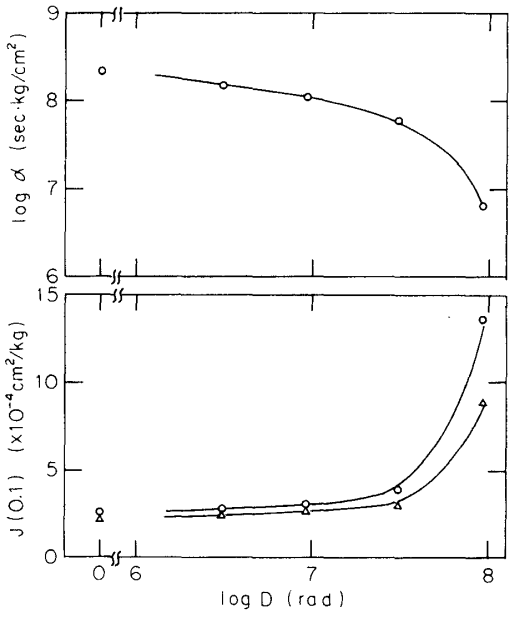


Fig. 9. Creep compliance J(0.1) and slope in J(t)−t curve α of Hinoki as a function of log D. (△: 20°C, ○: 50°C)

at time 0.1 min and the slope α at 100 min in **J**(t)−t curves for Hinoki wood as a function of dosage. **J**(0.1) value remains almost unchanged up to 1×10^7 rad, but starts to increase rapidly at about 3×10^7 rad and this tendency is similar to that of crystallinity. On the other hand, α which is a measure of creep rate, decreases with increasing dosage.

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