

# Correlation of Various Flow Indices

---

(出版者 / Publisher)

法政大学工学部

(雑誌名 / Journal or Publication Title)

法政大学工学部研究集報 / 法政大学工学部研究集報

(巻 / Volume)

14

(開始ページ / Start Page)

25

(終了ページ / End Page)

32

(発行年 / Year)

1978-03

(URL)

<https://doi.org/10.15002/00004018>

# Correlation of Various Flow Indices

Masayuki KASAJIMA\*

## Abstract

In the steady flow and the dynamic flow, correlation of various flow indices found from the flow characteristics in which the methods of expression are different were analyzed theoretically by using the new indicating method for the flow index. The non-Newtonian fluid models having exponential terms were applied to the flow characteristics. In the steady flow, relationship between the flow index obtained from the viscosity  $\eta$ -shear rate  $\dot{\gamma}$  flow characteristics and the flow index obtained from the shear stress  $\tau$ - $\dot{\gamma}$  flow characteristics is a linear function. The relationship between the flow index obtained from the  $\eta$ - $\tau$  flow characteristics and the flow index obtained from the  $\tau$ - $\dot{\gamma}$  (or the  $\eta$ - $\dot{\gamma}$ ) flow characteristics is a fractional function. When the ratio of flow index  $n$  to the  $\dot{\gamma}$  and the ratio of flow index  $m$  to  $\dot{\gamma}$  is expressed with logarithm, respectively, in the form of differential coefficient, the former is double the number of the latter.

## 1. Introduction

In non-Newtonian fluid engineering, the power law equation is one of the most useful flow equations. The flow index, that is, the exponential term in the power law equation, is a quantitative value expressing the fluidity. This flow index is the so-called structural viscosity index<sup>1)</sup>. It is known<sup>2)</sup> generally that the material having a higher cohesive energy has a higher structural viscosity index. Investigations by Oyanagi<sup>3)</sup> show that ordinarily the non-Newtonian flow index  $n$  (represented by the exponential term in the power law equation, having a constant) is larger for the resin having a higher compressibility. The viscosity of resin having a large value of  $n$  depends highly on the shear rate, because the value of  $n$  is a constant.

The steady flow characteristics can be represented by, for instance, mutually combining with shear stress  $\tau$ , shear rate  $\dot{\gamma}$  and viscosity  $\eta$ . The flow indices can be obtained from these flow characteristics. However, by the method of combining mutually  $\tau$ ,  $\dot{\gamma}$  and  $\eta$  gives a flow index having a different value. Therefore, it is significant that under different expression method of the flow characteristics, correlation of various flow indices obtained therefrom are become clear, and that the flow indices are indicated with the convenient method at engineering treatment. If these matters may be made clear, comparison of flow characteristics having different method of expression can be done easily, and the job of measuring the data may be decreased.

In this paper correlation of various flow indices, obtained respectively from the various characteristics of steady flow and dynamic flow, are discussed.

---

\* Department of Mechanical Engineering

## 2. Flow Index in Steady Flow

### 2.1 Flow index $n$

In a steady flow, the relationship of the shear stress  $\tau$ , the shear rate  $\dot{\gamma}$  and the viscosity  $\eta$  is expressed by the following equation.

$$\tau(\dot{\gamma},) = \eta(\dot{\gamma},) \cdot \dot{\gamma} \quad (1)$$

The following equation is obtained by differentiating Eq. (1).

$$\frac{\partial \eta}{\partial \dot{\gamma}} = \frac{1}{\dot{\gamma}} \left( \frac{\partial \tau}{\partial \dot{\gamma}} - \frac{\tau}{\dot{\gamma}} \right) \quad (2)$$

From Eq. (2),

$$\frac{\partial \ln \eta}{\partial \ln \dot{\gamma}} = \frac{\partial \ln \tau}{\partial \ln \dot{\gamma}} - 1 \quad (3)$$

is induced. And from Eq. (1),

$$\frac{\partial \eta}{\partial \tau} = \frac{1}{\dot{\gamma}} \left( 1 - \eta \frac{\partial \dot{\gamma}}{\partial \tau} \right) \quad (4)$$

is induced. The following equation is obtained from Eq. (4).

$$\frac{\partial \ln \eta}{\partial \ln \tau} = 1 - \frac{\partial \ln \dot{\gamma}}{\partial \ln \tau} \quad (5)$$

Flow indices  $n$ , obtained from the flow characteristics such as the  $\tau$ - $\dot{\gamma}$ ,  $\eta$ - $\dot{\gamma}$  and  $\eta$ - $\tau$ , are defined and indicated<sup>4)</sup> respectively, by using the new indicating method for the flow index, with the subscript as follows;

$$n_{\tau\dot{\gamma}} = \frac{\partial \ln \tau}{\partial \ln \dot{\gamma}} \quad (6)$$

$$n_{\eta\dot{\gamma}} = \frac{\partial \ln \eta}{\partial \ln \dot{\gamma}} \quad (7)$$

$$n_{\eta\tau} = \frac{\partial \ln \eta}{\partial \ln \tau} \quad (8)$$

The  $n_{\tau\dot{\gamma}}$  indicated by Eq. (6) agrees with the flow index  $n$  of  $n$ -power law<sup>5)</sup>. However, care should be taken that  $n$  of the  $n$ -power law is a constant. The  $n_{\tau\dot{\gamma}}$  in this paper is not constant and is expressed by generalized formula as a function of other influence factor.

Substituting Eqs. (6)~(8) into Eqs. (3) and (5) one obtains,

$$n_{\eta\dot{\gamma}} = n_{\tau\dot{\gamma}} - 1 \quad (9)$$

$$n_{\eta\tau} = 1 - \frac{1}{n_{\tau\dot{\gamma}}} \quad (10)$$

$$= \frac{n_{\eta\dot{\gamma}}}{n_{\eta\dot{\gamma}} + 1} \quad (11)$$

Eqs. (9)~(11) may be applied to every kind of fluid, for instance, pseudoplastic fluid, dilatant fluid, etc.. And these equations are not related to the respective functions of the flow indices.

The flow index corresponding to  $n_{\tau\dot{\gamma}}$  is used generally. However, the case where the

flow index  $n_{\eta\dot{\gamma}}$  is necessitated also occurs for example, in the case of the flow in the mold of the injection molding machine, etc.. Average fluidity, used in the case where the flow in the mold is investigated, is expressed<sup>6)</sup> by Eqs. (12) and (13).

$$\text{Average fluidity} = \alpha_{STV} \times M(\bar{\dot{\gamma}}, \bar{T}) \quad (12)$$

$$\alpha_{STV} = \frac{1}{\eta_0} \frac{(\partial \ln \eta) / (\partial \ln \dot{\gamma})}{(\partial \ln \eta) / \{\partial (1/T)\}} \quad (13)$$

where subscript *STV* of  $\alpha$  indicates capital letters of the shear rate, the temperature and the viscosity.  $M$  means the moldability<sup>7)</sup>.  $\eta_0$  is viscosity at the shear rate  $10 \text{ sec}^{-1}$ .  $T$ : temperature,  $\bar{\dot{\gamma}}$ : average shear rate,  $\bar{T}$ : average temperature. The  $\alpha_{STV}$  is the inherent value for the material itself and is independent of the shape of the mold etc.. The numerator of the right term of Eq. (13) corresponds to the flow index expressed by Eq. (7). Preferably a capillary rheometer is used for an experiment to obtain  $\alpha_{STV}$ , because, the flow in the capillary rheometer is analogous to the flow in the injection molding machine. Data obtained by the rheometer are usually  $\tau-\dot{\gamma}$  flow characteristics. Inducing the flow index from this  $\tau-\dot{\gamma}$  flow characteristics, the  $\alpha_{STV}$  can be found from the value of flow index and Eqs. (7), (9) and (13).

With the exception of the above sample case, Eqs. (9)~(11) are applied to the data of flow characteristics in which the method of expression is different to facilitate comparing the data. The applicable regions of the flow index for the pseudoplastic fluid are as follows,

$$0 < n_{\tau\dot{\gamma}} \leq 1 \quad (14)$$

$$-1 < n_{\eta\dot{\gamma}} \leq 0 \quad (15)$$

$$n_{\eta\tau} \leq 0 \quad (16)$$

The case of  $n_{\tau\dot{\gamma}}=1$  means the Newtonian fluid. For the non-Newtonian fluid the flow index  $n$  can be expressed<sup>4),9),10)</sup> by the equations which are a function of the shear rate etc.. A formula of  $n_{\tau\dot{\gamma}}$  is expressed as follows,

$$n_{\tau\dot{\gamma}}(\dot{\gamma}, a_T, a_P) = \sum_{j=0}^4 (\alpha_{\tau\dot{\gamma}})_{oj} \{ \ln(a_T a_P \dot{\gamma}) \}^j \quad (17)$$

where  $a_T$ : the temperature shift factor,  $a_P$ : the pressure shift factor,  $\alpha$ : a constant, subscript  $\tau\dot{\gamma}$ : a value in  $\tau-\dot{\gamma}$  flow curve, subscript  $o$ : a value at the state of reference temperature and reference hydrostatic pressure. It is possible<sup>10)~16)</sup> that, a relation between  $a_T$  shown in Eq. (17) and the temperature, and a relation between  $a_P$  shown in Eq. (17) and the hydrostatic pressure, can be decided respectively.

There is Eq. (18)<sup>17),18)</sup> for a model in which the flow index  $n$  is a function of the shear stress.

$$n_{\tau\dot{\gamma}}(\tau) = \sum_{j=0}^4 (\beta_{\tau\dot{\gamma}})_{oj} (\ln \tau)^j \quad (18)$$

where  $\beta$  means a constant. In addition, the formulae of Eqs. (17) and (18) are independent of each other. When  $n_{\tau\dot{\gamma}}$  is expressed by Eq. (17) and Eq. (18),  $n_{\eta\dot{\gamma}}$  and  $n_{\eta\tau}$  are found easily by using Eqs. (9)~(11), (17) and (18).

Let's investigate the relationship between the flow index and the shift factor. The

relationship between  $n_{\tau\dot{\gamma}}$  and the temperature shift factor  $a_T$ , and the relationship between  $n_{\tau\dot{\gamma}}$  and the pressure shift factor  $a_P$ , are expressed respectively by the following equations, using by the model equation indicated in Eq. (17).

$$\frac{\partial n_{\tau\dot{\gamma}}}{\partial \ln \dot{\gamma}} = \frac{\partial n_{\tau\dot{\gamma}}}{\partial \ln a_T} \quad (19)$$

$$\frac{\partial n_{\tau\dot{\gamma}}}{\partial \ln \dot{\gamma}} = \frac{\partial n_{\tau\dot{\gamma}}}{\partial \ln a_P} \quad (20)$$

Eqs. (19) and (20) denote that, at the same value of the flow index, respective inclinations  $n_{\tau\dot{\gamma}} - \ln \dot{\gamma}$  curve and  $n_{\tau\dot{\gamma}} - \ln a_T$  curve, respective inclinations of  $n_{\tau\dot{\gamma}} - \ln \dot{\gamma}$  curve and  $n_{\tau\dot{\gamma}} - \ln a_P$  curve, have the same value respectively.

## 2.2 Flow index $m$

Formulae for the non-Newtonian fluid model<sup>(8)~(10), (19)</sup> in which the flow index  $n_{\tau\dot{\gamma}}$  is denoted by Eq. (17) are expressed as follows,

$$\tau = \tau_o^\circ \left( \frac{a_T a_P \dot{\gamma}}{\dot{\gamma}_o^\circ} \right)^{m_{\tau\dot{\gamma}}(\dot{\gamma}, \dot{\gamma}_o^\circ, a_T, a_P)} \quad (21)$$

$$\eta = a_T a_P \eta_o^\circ \left( \frac{a_T a_P \dot{\gamma}}{\dot{\gamma}_o^\circ} \right)^{m_{\eta\dot{\gamma}}(\dot{\gamma}, \dot{\gamma}_o^\circ, a_T, a_P)} \quad (22)$$

$$\eta = a_T a_P \eta_o^\circ \left( \frac{\tau}{\tau_o^\circ} \right)^{m_{\eta\tau}(\tau, \tau_o^\circ)} \quad (23)$$

$$m_{\tau\dot{\gamma}}(\dot{\gamma}, \dot{\gamma}_o^\circ, a_T, a_P) = \sum_{i=0}^j \left\{ \frac{(\alpha_{\tau\dot{\gamma}})_o}{j+1} \sum_{k=0}^j \left\{ \ln(a_T a_P \dot{\gamma}) \right\}^{j-k} (\ln \dot{\gamma}_o^\circ)^k \right\} \quad (24)$$

where subscript  $\circ$ : a value at reference temperature and reference hydrostatic pressure, superscript  $\circ$ : a value at standard state. Correlation of various flow indices  $m$  in the exponential equations indicated by Eqs. (21)~(23), from Eqs. (1) and (21)~(23), are expressed as follows,

$$m_{\eta\dot{\gamma}} = m_{\tau\dot{\gamma}} - 1 \quad (25)$$

$$m_{\eta\tau} = 1 - \frac{1}{m_{\tau\dot{\gamma}}} \quad (26)$$

$$= \frac{m_{\eta\dot{\gamma}}}{m_{\tau\dot{\gamma}} + 1} \quad (27)$$

Eqs. (25)~(27) make clear that the relationship between  $m_{\eta\dot{\gamma}}$  and  $m_{\tau\dot{\gamma}}$  is a linear function, the relationship  $m_{\eta\tau}$  relative to  $m_{\tau\dot{\gamma}}$  and  $m_{\eta\dot{\gamma}}$  is a relation of fractional function. Eqs. (21)~(27) may be applied to pseudoplastic fluid and dilatant fluid etc.. The region of the flow index  $m$  differs with the kind of fluid. The applicable regions of the flow index  $m$  for the pseudoplastic fluid are as follows,

$$0 < m_{\tau\dot{\gamma}} \leq 1 \quad (28)$$

$$-1 < m_{\eta\dot{\gamma}} \leq 0 \quad (29)$$

$$m_{\eta\tau} \leq 0 \quad (30)$$

Let's investigate the relationship between the flow index  $m_{\tau\dot{\gamma}}$  and the shift factors. The following equations are obtained from Eqs. (21)~(24).

$$\frac{\partial m_{\tau\dot{\gamma}}}{\partial \ln \dot{\gamma}} = \frac{\partial m_{\tau\dot{\gamma}}}{\partial \ln a_T} \tag{31}$$

$$\frac{\partial m_{\tau\dot{\gamma}}}{\partial \ln \dot{\gamma}} = \frac{\partial m_{\tau\dot{\gamma}}}{\partial \ln a_P} \tag{32}$$

From Eqs. (17) and (24), Eq. (33) which expresses a relationship between the flow index  $n_{\tau\dot{\gamma}}$  and  $m_{\tau\dot{\gamma}}$ , is obtained.

$$\frac{\partial n_{\tau\dot{\gamma}}}{\partial \ln \dot{\gamma}} = 2 \frac{\partial m_{\tau\dot{\gamma}}}{\partial \ln \dot{\gamma}} \tag{33}$$

Viewing the figures, Eq. (33) denotes that the number of inclinations of  $n_{\tau\dot{\gamma}} - \ln \dot{\gamma}$  curve is double that inclination of  $m_{\tau\dot{\gamma}} - \ln \dot{\gamma}$  curve, at the location where  $n_{\tau\dot{\gamma}}$  curve and  $m_{\tau\dot{\gamma}}$  curve intersect each other.

### 3. Flow Index in Dynamic Flow

The constitutive equation in the dynamic flow can be expressed<sup>19)</sup> by an exponential formula. The models for dynamic rigidity  $G'$ -angular frequency  $\omega$  flow characteristics are indicated as follows,

$$G' = G'_0 \left( \frac{a_T a_P \omega}{\omega_0} \right)^{m'_{G'\omega}(\omega, \omega_0, a_T, a_P)} \tag{34}$$

$$m'_{G'\omega}(\omega, \omega_0, a_T, a_P) = \sum_{j=0}^i \left[ \frac{(\alpha'_{G'\omega})_{0j}}{j+1} \sum_{k=0}^j \left\{ \ln(a_T a_P \omega) \right\}^{j-k} (\ln \omega_0)^k \right] \tag{35}$$

$$n'_{G'\omega}(\omega, a_T, a_P) = \sum_{j=0}^i (\alpha'_{G'\omega})_{0j} \{ \ln(a_T a_P \omega) \}^j \tag{36}$$

where  $\alpha'$ : a constant, superscript ': in the dynamic flow, subscript  $G'\omega$ : a value in  $G' - \omega$  curve. The procedure to obtain the  $G' - \omega$  flow characteristics is generally different from the procedure to obtain the  $\eta' - \omega$  flow characteristics. Relationship among  $G'$ ,  $\omega$  and  $\eta'$  can not be represented by such as Eq. (1) in a steady flow. Therefore, fluid models for the  $\eta' - \omega$  flow characteristics should be denoted by different fluid models from the  $G' - \omega$  flow characteristics. The fluid models for the  $\eta' - \omega$  are indicated as follows,

$$\eta' = a_T a_P \eta'_0 \left( \frac{a_T a_P \omega}{\omega_0} \right)^{m'_{\eta'\omega}(\omega, \omega_0, a_T, a_P)} \tag{37}$$

$$m'_{\eta'\omega}(\omega, \omega_0, a_T, a_P) = \sum_{j=0}^i \left[ \frac{(\alpha'_{\eta'\omega})_{0j}}{j+1} \sum_{k=0}^j \left\{ \ln(a_T a_P \omega) \right\}^{j-k} (\ln \omega_0)^k \right] \tag{38}$$

$$n'_{\eta'\omega}(\omega, a_T, a_P) = \sum_{j=0}^i (\alpha'_{\eta'\omega})_{0j} \{ \ln(a_T a_P \omega) \}^j \tag{39}$$

The relationship of various dynamic flow indices  $n'$  can not be represented by relations as Eqs. (9)~(11). The relationship of various dynamic flow indices  $m'$  can not be represented by such relations as Eqs. (25)~(27). However, Eq. (40) which indicates the relation between  $n'_{G'\omega}$  and  $m'_{G'\omega}$ , is induced from Eqs. (35) and (36). From Eqs. (38) and (39),  $n'_{\eta'\omega} - m'_{\eta'\omega}$  relation is obtained as the following Eq. (41).

$$\frac{\partial n'_{G'\omega}}{\partial \ln \omega} = 2 \frac{\partial m'_{G'\omega}}{\partial \ln \omega} \tag{40}$$

$$\frac{\partial n'_{\tau, \omega}}{\partial \ln \omega} = 2 \frac{\partial m'_{\tau, \omega}}{\partial \ln \omega} \quad (41)$$

In the case where the fluid models in the dynamic flow are represented by Eqs. (34)~(39), relationships between the dynamic flow index and the shift factor may be expressed by the formulae, which are replaced  $n_{\tau_j}$  with  $n'_{\tau, \omega}$ ,  $\dot{\gamma}$  with  $\omega$  in Eqs. (19) and (20), which are replaced  $m_{\tau_j}$  with  $m'_{\tau, \omega}$ ,  $\dot{\gamma}$  with  $\omega$  in Eqs. (31) and (32), respectively.

#### 4. Various Flow Index Curves

Using the results analyzed in the previous section, various flow index curves are shown schematically in Fig. 1 which is a sample in a case where the equations of non-Newtonian

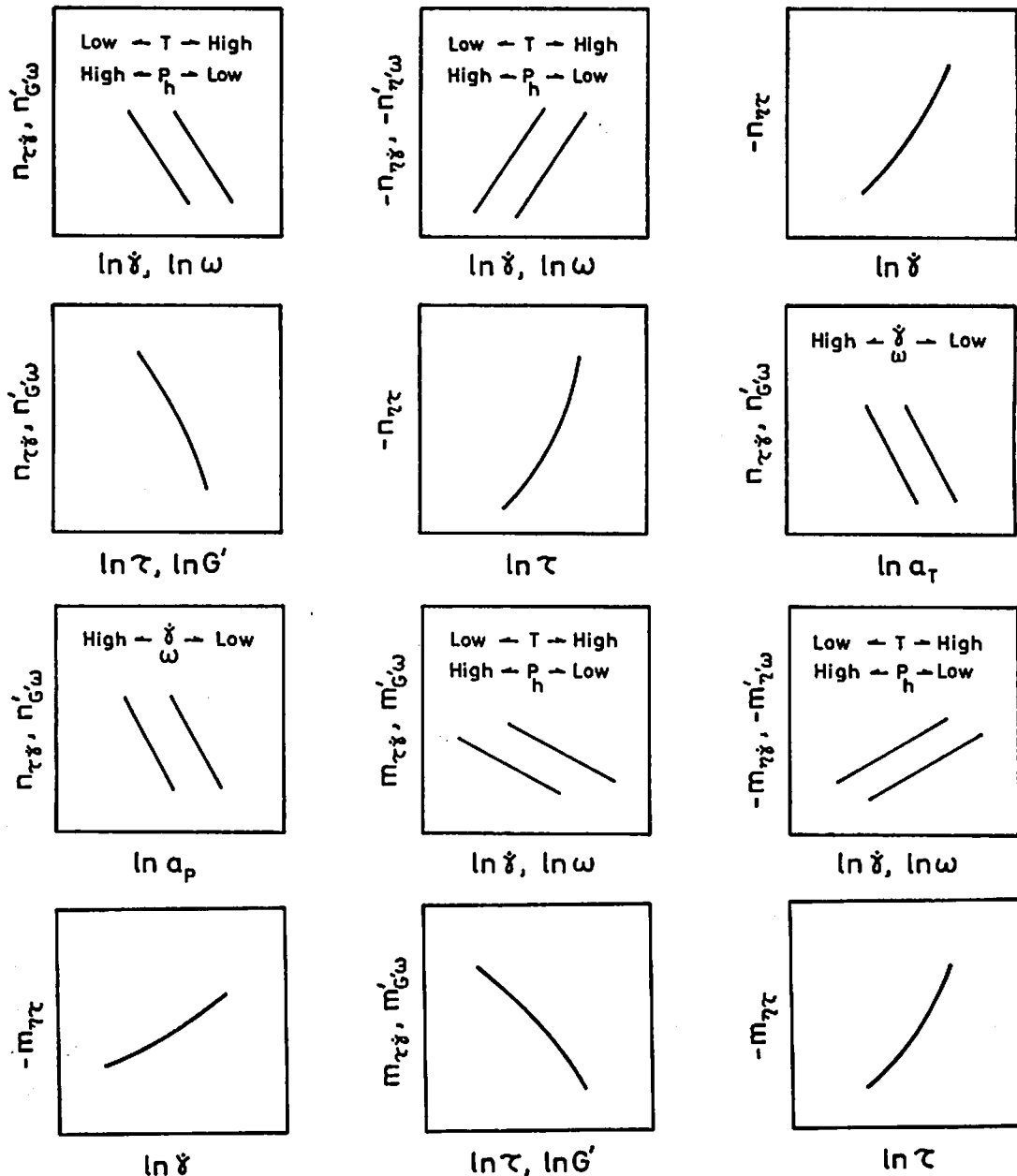


Fig. 1 Schematic illustration of various flow index curves

fluid model (indicated in the previous section) at  $i=1$  are applied to the pseudoplastic fluid. In practical flow, there are, too, the cases where " $i$ " has a larger value than unity. Two symbols written on the ordinate in Fig. 1 indicate respectively the values in steady flow and in dynamic flow. Hence, in reading the figure, various symbols should be read by corresponding to respective flow systems.

## 5. Conclusion

The correlations of various flow indices found from the different various flow characteristics in steady flow and dynamic flow were investigated by using the new indicating method for the flow index. The followings were made clear.

1) In the steady flow, for every kind of the shape of the function (e.g., function of the shear rate etc.) of the flow indices  $n$  and  $m$ , the relationship between the flow index obtained from the viscosity-shear rate flow characteristics, and the flow index obtained from the shear stress-shear rate flow characteristics is a linear function. The relationship between the flow index obtained from the viscosity-shear stress flow characteristics, and the flow index obtained from the shear stress-shear rate (or the viscosity-shear rate) flow characteristics, is a fractional function. These relationships for the flow index  $n$  are valid for every kind of fluid. The above flow index  $m$  relationships are valid for any fluid to which the non-Newtonian fluid model of  $m$ -power law may be applied.

2) In dynamic flow, the relationship of various dynamic flow indices  $n'$  and the relationship of various dynamic flow indices  $m'$  can not be represented by such relations as the various flow indices in the steady flow, respectively.

3) In the flow index-shear rate (indicated with logarithm) curves, the number of inclinations of  $n$  curve is double the number of the inclination of  $m$  curve at the point where  $n$  and  $m$  curves intersect each other.

These relationships make a comparative study of the flow characteristics in which the methods of expression are different, easy and also then may be used effectively to estimate flow characteristics other than measured values.

## Acknowledgements

The author expresses his thanks to Professor Suganuma, Professor Kunii, Professor Mori of University of Tokyo and Professor Ito of Hosei University whose advice made this study possible.

## Nomenclature

- $a_p$  = pressure shift factor
- $a_r$  = temperature shift factor
- $G'$  = dynamic rigidity
- $M$  = moldability defined in Eq. (12)
- $m$  = flow index
- $n$  = flow index
- $P_h$  = hydrostatic pressure



- $T$  =temperature  
 $\bar{T}$  =average temperature  
 $\alpha$  =constant  
 $\alpha_{STV}$  =defined in Eq. (13)  
 $\beta$  =constant  
 $\dot{\gamma}$  =shear rate  
 $\bar{\dot{\gamma}}$  =average shear rate  
 $\eta$  =viscosity in steady flow  
 $\eta'$  =dynamic viscosity  
 $\eta_0$  =viscosity at  $\dot{\gamma}=10 \text{ sec}^{-1}$  in Eq. (13)  
 $\tau$  =shear stress  
 $\omega$  =angular frequency  
 <Superscripts>  
 $\circ$  =standard state  
 $'$  =dynamic  
 <Subscripts>  
 $\circ$  =reference point  
 $G'\omega$  =in flow curve  $G'-\omega$   
 $\eta\dot{\gamma}$  =in flow curve  $\eta-\dot{\gamma}$   
 $\eta\tau$  =in flow curve  $\eta-\tau$   
 $\eta'\omega$  =in flow curve  $\eta'-\omega$   
 $\tau\dot{\gamma}$  =in flow curve  $\tau-\dot{\gamma}$

### References

- 1) Polymer Jiten Henshu Iinkai: "Dictionary of Polymer Technology", p.130, Taiseisha (1970).
- 2) Uno, T.: "Poly-Enka-Vinyl, II", p.232, Asakura-shoten (1966).
- 3) Oyanagi, Y. and A. Yamaguchi: 22nd Kobunshi Toronkai Maezuri, III-357 (1973).
- 4) Kasajima, M. and A. Sugauma: Chukaren Shukitaikai Koenyokoshu, 193 (1975).
- 5) McKelvey, J.M.: "Polymer Processing", p.31, John Wiley (1962).
- 6) Ito, K.: Kikai no Kenkyu, 18, 11, 59 (1966).
- 7) Weir, F.E.: SPE Technical Paper, 9, 4, (1962).
- 8) Kasajima, M.: Mori Kenkyushitsu Danwakai Shiryo, Univ. of Tokyo (1974).
- 9) Kasajima, M., Y. Mori and A. Sugauma: *Kagaku Kogaku Ronbunshu*, 1, 137 (1975).
- 10) Kasajima, M., A. Sugauma and D. Kunii: 41st Kagaku Kogaku Kyokai Nenkai Yoshishu, 142 (1976).
- 11) Williams, M.L., R.F. Landle and J.D. Ferry: *J. Am. Chem. Soc.*, 77, 3701 (1955).
- 12) Arai, T.: *Kobunshi Kagaku*, 18, 292 (1961).
- 13) Horio, M., T. Fujii and S. Onogi: *J. Phys. Chem.*, 68, 778 (1964).
- 14) Ito, K.: Kikai no Kenkyu, 19, 3, 75 (1967).
- 15) Kasajima, M., A. Sugauma and D. Kunii: 25th Kobunshi Gakkai Nenjitaikai Yokoshu, 468 (1976).
- 16) Hayashi, S.: "Rheology", p.157, Kodansha (1973).
- 17) Kasajima, M., A. Sugauma and D. Kunii: 3rd Nippon Rheology Gakkai Nenkai Yoshishu, 17 (1976).
- 18) Kasajima, M., A. Sugauma and D. Kunii: 25th Kobunshi Gakkai Nenjitaikai Yokoshu, 470 (1976).
- 19) Kasajima, M., A. Sugauma and D. Kunii: 25th Kobunshi Gakkai Nenjitaikai Yokoshu, 468 (1976). *ibid*, 469 (1976).