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| 著者 | SHIRAKAWA Yuki, NUMAKURA Ken'ichi |
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# On Young's Modulus and Grain Size in Nickel-Copper Alloys*

Yûki SHIRAKAWA and Ken'ichi NUMAKURA
*The Research Institute for Iron, Steel and Other Metals*
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**Synopsis**

Young's modulus and the grain size have been measured with 10 kinds of polycrystalline ferromagnetic nickel-copper alloys annealed at 700°, 800°, 900°, 1000° and 1100°C. The following empirical relation has been found between Young's modulus $E$ and the average area of crystal grains $S$:

$$E = E_0 - B \log S,$$

where $E_0$ and $B$ are constants. Young's modulus vs. composition curves for alloys with the same grain size were not always similar to one another, but every curve showed a minimum. The composition corresponding to the minimum shifted toward the nickel side as the grain size became large. With the addition of copper, Young's modulus of nickel with comparatively small grains increased, whereas that with comparatively large grains decreased. These results suggest that the complication of the so-far as observed Young's modulus vs. composition curves for nickel-copper alloys may be due partly to the difference in the grain size of the individual specimens. Finally, the measured results were compared with those calculated by the formula derived by Voigt and Reuss.

## I. Introduction

young's modulus of polycrystalline ferromagnetic nickel-copper alloys have been measured by many inventigators(1), but their results are very different from one anoter as shown in Fig. 1. Many physicists have given their attentions to this disagreement, but it is certainly unkown to what facts it is due. The present authors also have taken previously an intrest in this problem, and so, preparing the single crystals of these alloys, made clear the dependene of the orientation on Young's modulus(2).

In this paper, the variation of Young's modulus due to grain size was explained, by which one factor of the inagreement of previous results could be clarified.

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* The 904 th report of the Research Institute for Iron, Steel and Other Metals. Published in Japanese in Nippon Kinzoku Gakkai-shi (J. Japan Inst. Metals), **19** (1955), 546.

(1) Z. Nishiyama, Sci. Rep. Tohoku Univ., **18** (1929), 359; K. Nakamura, **25** (1936), 415; S. Aoyama and T. Fukuroi, Bull. Inst. Phys. Chem. Res., **20** (1941), 809; M. Yamamoto, Nippon Kinzoku Gakkai-shi (J. Japan Inst. Metals), **6** (1942), 249; Sci. Rep. RITU, **A6** (1954), 446; H. Masumoto and H. Saito, Nippon Kinzoku Gakkai-shi, **8** (1944), 49; W. Köster and W. Rauscher, Z. Metallkde., **39** (1948), 111, T. Fukuroi rnd Y. Shibuya, Sci. Rep. RITU, **A2** (1950), 748; J.S. Kouvelites and L.W. Mckeeham, Phys. Rev., **86** (1952), 898. S. Umekawa, Nippon Kinzoku Gakkaishi, **18** (1954), 387.

(2) Y. Shirakawa and K. Numakura, Sci. Rep. RITU, **A10** (1957), 51, Nippon Kinzoku Gakkai-shi, **19** (1955), 99.

## II. Specimens, apparatus for experiment and method of measurement

With electrolytic nickel and copper of high purity, 10 kinds of specimens were prepared by using a high frequency induction furnace. After being forged, they were shaved into plates, 10 cm in length, 5 mm in width and 1 mm in thickness, and finally filed up.

Then, the specimens were annealed at 700°, 800°, 1000° and 1100°C for 1 hour in high vacuum in order to prepare the specimens having various grain sizes. The chemical analyses of specimens are shown in Table 1. Young's modulus was measured by usual method of bending. The distance between the knife edges was about 8 cm, and the distance between two mirrors was about 9 cm, and the distance between scale and facing mirror was about 100 cm.

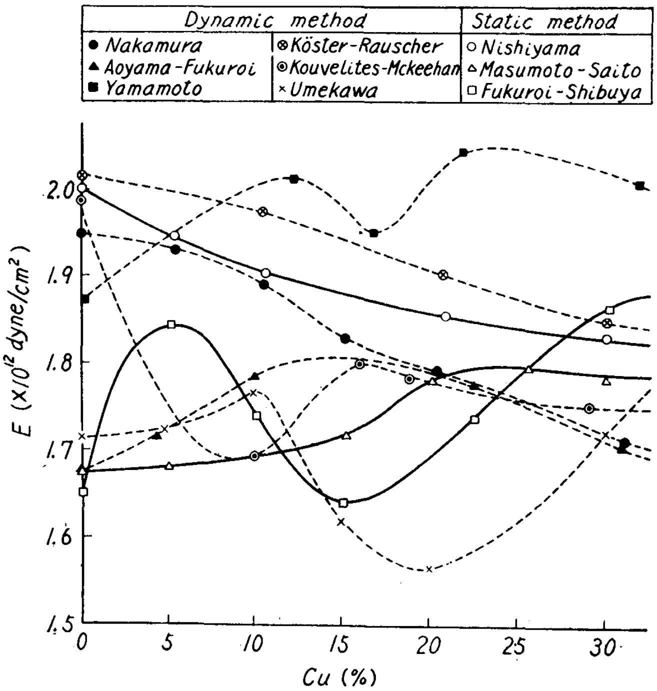


Fig. 1. Young's modulus vs. composition curves for Ni-Cu alloys obtained by various investigators.

A paper dish of about 2 g in weight was always hung at the center of the specimen, and the maximum stress at the neutral axis by this dish was about 5 kg/cm² (=$4.9\times10^6$ dyne/cm²), which was negligibly small for the present purpose.

The weight on the dish was 5, 10 and 20 g. The results shown in Table 1 are the average values of these measurements. The highest maximum stress at the

Table 1. Chemical analysis, Young's modulus, $E$ and average area of crystal grains, $S$ for various heat treatments in Ni-Cu alloys.

| Composition (%) | | | 700 (°C) | | 800 (°C) | | 900 (°C) | | 1000 (°C) | | 1100 (°C) | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Cu | C | Mn | $E$ ($\times10^{12}$ dyne/cm²) | $S$ ($\times10^{-4}$ mm²) | $E$ ($\times10^{12}$ dyne/cm²) | $S$ ($\times10^{-4}$ mm²) | $E$ ($\times10^{12}$ dyne/cm²) | $S$ ($\times10^{-4}$ mm²) | $E$ ($\times10^{12}$ dyne/cm²) | $S$ ($\times10^{-4}$ mm²) | $E$ ($\times10^{12}$ dyne/cm²) | $S$ ($\times10^{-4}$ mm²) |
| — | 0.008 | 0.093 | 1.70 | 28 | 1.69 | 37 | 1.68 | 53 | 1.66 | 97 | 1.65 | 109 |
| 3.15 | 0.010 | 0.053 | 1.72 | 25 | 1.70 | 27 | 1.69 | 40 | 1.67 | 55 | 1.64 | 100 |
| 4.95 | 0.009 | 0.090 | 1.72 | 20 | 1.69 | 25 | 1.69 | 30 | 1.63 | 57 | 1.60 | 86 |
| 8.09 | 0.014 | 0.073 | 1.75 | 10.4 | 1.70 | 20 | 1.68 | 27 | 1.61 | 66 | 1.58 | 110 |
| 11.96 | 0.010 | 0.130 | 1.70 | 17 | 1.68 | 21 | 1.67 | 31 | 1.64 | 44 | 1.63 | 61 |
| 14.54 | 0.012 | 0.087 | 1.75 | 17 | 1.72 | 27 | 1.70 | 43 | 1.70 | 44 | 1.69 | 53 |
| 17.59 | 0.015 | 0.160 | 1.82 | 9.6 | 1.80 | 13 | 1.77 | 35 | 1.75 | 40 | 1.74 | 53.4 |
| 21.53 | 0.009 | 0.150 | 1.84 | 10 | 1.82 | 12 | 1.81 | 17 | 1.80 | 20 | 1.78 | 35 |
| 24.61 | 0.010 | 0.043 | 1.78 | 21 | 1.75 | 42 | 1.73 | 62 | 1.72 | 70 | 1.70 | 118 |
| 29.87 | 0.014 | 0.067 | 1.80 | 9 | 1.79 | 11 | 1.74 | 24 | 1.74 | 25 | 1.72 | 32 |

neutral axis by the largest weight was about 53 kg/cm² ($=52\times10^6$ dyne/cm²). This value was only about 0.125 of the elastic limit even for copper having the largest probability of plastic flow. In addition, the experiment was repeated when the reading on a scale does not come back to the reading before loading, leaving the weight out from the dish. The average grain size was determined by the following equation of Zemmer:

$$S = A/(n_1 + 0.6\,n_2) \cdot m^2, \qquad \cdots\cdots\cdots\cdots(1)$$

where $S$ is the average area of crystal grains, $A$ the area of the field of vision in microscope, $n_1$ the number of grains in the circle, $n_2$ the number of grains cut by the circumference of the circle and $m$ the magnification of microscope.

## III. Experimental results and the consideration

1. Heating temperature and grain size

The grain size of specimen annealed respectively at 700°, 800°, 900°, 1000° and 1100°C is shown in Table 1. Though the specimens were subjected to the same heat treatment, the grain size was different with different copper concentrations. The $S$ vs. copper concentration curves for the annealing temperature of 700°, 900° and 1100°C are shown in Fig. 2. The vertical axis $S$ ($\times10^{-4}$mm²) in this figure is of a logarithmic scale. As shown in the figure, the tendencies of curves for

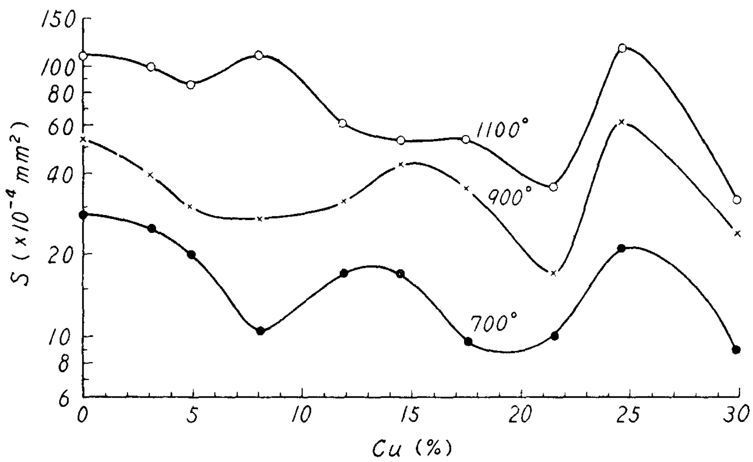


Fig. 2. Relation between average area of crystal grains, $S$ and Cu composition for Ni-Cu alloys annealed at 700°, 900° and 1100°C.

respective temperatures were almost similar to one another, but the intrevals between curves were very different with different copper concentrations. It may be due to the condition of the specimens before heating, that is, the circumstances of preparation, or the impurities and the degree of working. But, the difference in the rate of growth and the relaxation time in the recrystallization may be more essential.*

* These experiments are going on in our laboratory.

2. Young's modulus and grain size

Young's modulus in the neighbourhood of grain boundaries may be larger than in the interior of crystal grain, because the former has more lattice strain than the latter. Therefore, Young's modulus of specimens subjected to various heat treatments, that is, with the different grain sizes was measured and is shown in Table 1. As shown in Table 1, Young's modulus is differerent with different grain sizes.

When Young's modulus E is plotted aginst the logarithm of the average area of crystal grain S, the measuring points almost lay on a straight line. For example,

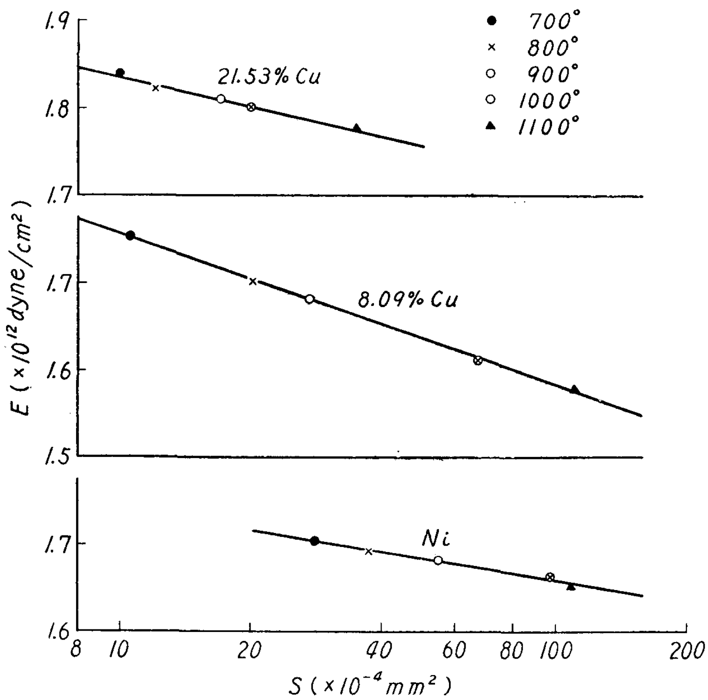


Fig. 3. Relation between Young's modulus, $E$ and average area of crystal grains $S$.

those of nickel and alloys containing 8.09 and 21.53 per cent of copper are shown in Fig. 3. Therefore, within the present experimental range, the following equation was obtained:

$$E = E_0 - \mathrm{B} \log S. \qquad \cdots\cdots\cdots\cdots(2)$$

The grain size $S$ seems to have some limit in order that the above equation hold good, which, however, could not be ascertained.

If the straight lines of individual specimens are drawn as shown in Fig. 3 and Young's modulus with $S = 10$, 30 and $100 \times 10^{-4}$ mm$^2$ is plotted against copper concentration, the full lines in Fig. 4 will be obtained. As shown in the figure, the curve with $S = 10 \times 10^{-4}$ mm$^2$ showed a maximum at 5 per cent, a minimum at 11 per cent and another maximum at 20 per cent of copper. The copper concentration showing the first maximum and minimum decreases with the increase of grain size, and in the curve for $S = 100 \times 10^{-4}$ mm$^2$ the first maximum disappears and a minimum appears at 7 per cent of copper. However, the maximum point at

20 per cent of copper is invariable. It seems, therefore, that the final maximum is due to the transition from ferromagnetic to paramagnetic range, but that the first maximum and minimum in ferromagnetic range are unexplainable.

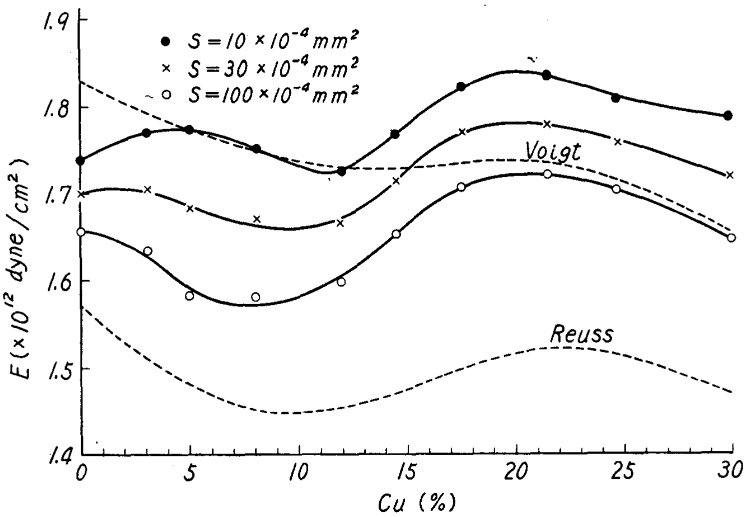


Fig. 4. Young's modulus vs. composition curves for Ni-Cu alloys. The full lines are Young's modulus of polycrystals with various grain sizes and dotted lines calculated from the formula derived by Voigt and Reuss using our results of single crystals.

3. Comparison with previous results

The previous result were obtained by the static method of bending and the dynamic method of vibration as shown in Eig. 1, but in every case, 4 kinds of tendencies were recognized, that is,

(A) the value of nickel simply decreases with the incease in copper content;
(B) the value of nickel increases at first until a maximum point, and then dcreases;
(C) the curve shows a minimum in the course of increasing;
(D) the value of nickel decreases at first until a minimum point, increases until a maximum point, and then decreases.

It is very difficult to account for these complicated aspect only by the present results, but it was found that E vs. copper concentration curves were not identical with one another as the grain size was not the same even at the same heat treatment, from which at least a part of complicated anomaly may be explained.

The heat treatments of specimens used by various investigators are shown in Table 2.

Now, the previous results in Fig. 1 will be speculated from the present results in Fig. 4. As the curves in Fig. 1 were obtained irrespective of grain size, if the values of specimens having different grain sizes are adopted, various curves will be obtained. However, provided that its effect is small, it is considered that the grain size is the smallest in the case of (A), smaller in the case of (B) and (C), and the largest in the case of (D).

The curve for the large grain size in Fig. 4 is similar to the tendency of curve obtained by Konvelites and Mckeehan[1], but their measuring points are insufficient

Table 2. Heat treatments of specimens used by various investigators.

| Method | Investigator | Temp. (°C) | Time (hr) | Atmo-sphere | Note |
|---|---|---|---|---|---|
| Dynamic | Nakamura (1936) | — | — | — | A |
| | Aoyama-Fukuroi (1941) | 900 | 6 | vac. | B |
| | Yamamoto (1942) | 850 | 3 | vac. | C |
| | Köster-Rauscher (1948) | — | — | — | A |
| | Kouvelites-Mckeehan (1952) | 700 | 3 | $H_2$ | D |
| | Umekawa (1954) | 850 | 0.5 | vac. | C |
| Static | Nishiyama (1929) | 900 | 2 | vac. | A |
| | Masumoto-Saito (1944) | 1000 | 1 | vac. | B |
| | Fukuroi-Shibuya (1950) | 900 | 1 | vac. | C |
| | Shirakawa-Numakura (1955) | 700∼1100 | 1 | vac. | D |

in the composition range in question. Furthermore, the curve for the large grain size was similar to that of [100] direction of single crystals obtained previously(2), so that the existence of the anisotropy was detected by X-ray photograph, but every specimen had not orientation worthy of special consideration.

4. Comparison with the results obtained with single crystals

Hitherto, the theoretical equations obtaining Young's modulus of a polycrystal from those of the single crystals have been proposed by Voigt and Reuss. Voigt(3) derived the following expression under the assumption that the strain is constant in all grains:

$$E = (C_{11}+2C_{12})\ (C_{11}-C_{12}+3C_{44})/(C_{11}+3C_{12}+C_{44}). \qquad \cdots\cdots\cdots\cdots(3)$$

On the contrary, Reuss(4) derived the following equation under the assumption that the stress is constant in all grains:

$$E = 5/(3S_{11}+2S_{12}+S_{44}). \qquad \cdots\cdots\cdots\cdots(4)$$

The curves of dotted lines shown in Fig. 4 are obtained by Eqs. (3) and (4) with Young's modulus of the single crystals obtained by the present authors. As shown in the figure, the experimental values are between the two values obtained by Eqs. of Voigt and of Reuss.

This agrees with what recently Pursey and Cox(5) made clear. Further, the tendencies of curves obtained by the theoretical equations are similar to that in the case of large grain size. On the other hand, the relation between Young's modulus of single crystals and that of polycrystal was derived more simply by the presnt authors, that is, denoting Young's modulus of polycrystal by $E$, and that for [i]-th direction of every crystallites by $E_i$, the following relation is obtained, provided that the effect of crystal grain boundary is negligible:

(3) W. Voigt, *Lehrbuch der Kristallphysik*, (1910).
(4) A. Reuss, Z. angew, Math. u. Mech., **9** (1929), 49.
(5) H. Pursey and H.L. Cox, Phil. Mag., **45** (1954), 295.

$$1/E = \sum_{i=1}^{j} 1/E_i \times P_i, \qquad \cdots\cdots\cdots\cdots(5)$$

were $P_i$ is the probability that the characteristic of [i]-th direction contributes to a special direction. (For instance, the direction that Young's modulus is measured). Assuming that individual crystallites are the sphere of the same size,

$$1/E = \sum_{i=1}^{j} 1/E_i \times k_i/\sigma, \qquad \cdots\cdots\cdots\cdots(6)$$

where $k_i$ is the number that [i]-th direction appears repeatedly over all directions, and $\sigma$ total sum of them. Young's moduli in principal axes $E_{[100]}$, $E_{[100]}$ and $E_{[111]}$ were obtained by the present authors so that only three directions of [100], [110] and [111] were selected as [i]-th direction. The number of directions equivalent

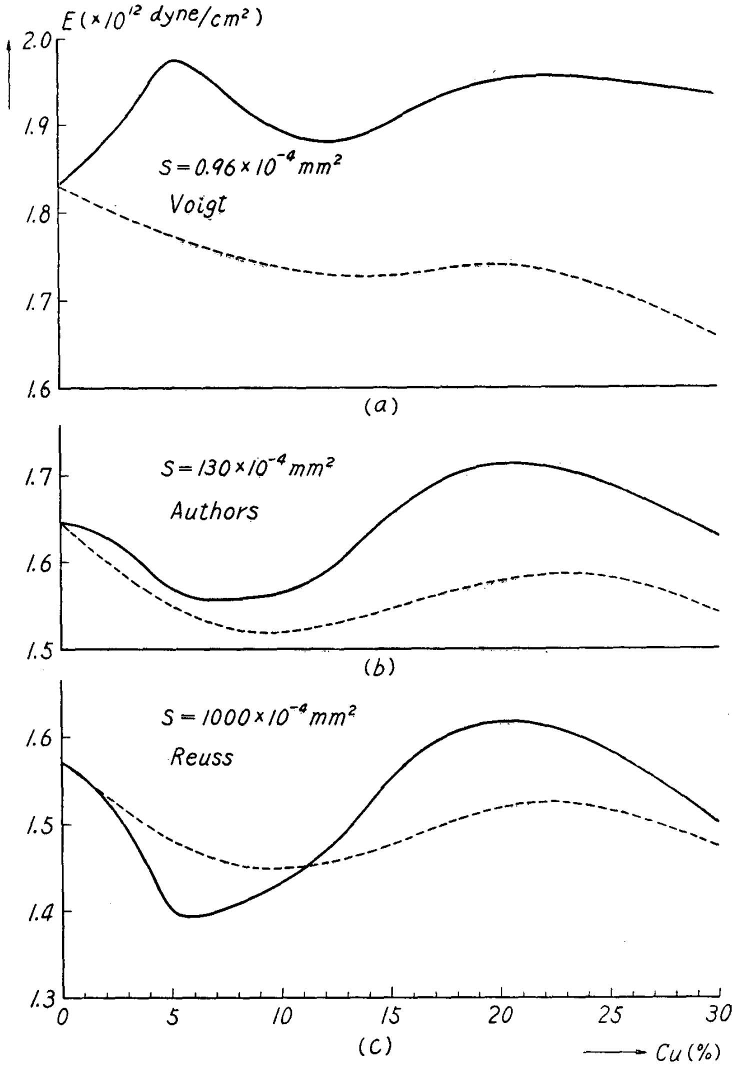


Fig. 5. Young's modulus vs. concentration curves for Ni-Cu alloys. The full lines in (a), (b), (c) are respectively the Young's modulus of polycrystal with 0.96, 130, $1000 \times 10^{-4}$ mm$^2$ grain sizes and the dotted lines calculated from the formulla derived by Voigt, authors and Reuss using our results of single crystals.

to [100], [110] and [111] over all directions are 6, 12 and 8, respectively. Therefore, using Eq. (6), the following formula will be derived:

$$1/E = 1/13\,(3/E_{[100]} + 6/E_{[110]} + 4/E_{[111]}). \qquad \cdots\cdots\cdots\cdots(7)$$

Now, Eq. (7) will be compared with Reuss's Eq. (4), by changing the latter into the form,

$$1/E = 1/5\,(2/E_{[100]} + 3/_{[111]}), \qquad \cdots\cdots\cdots\cdots(8)$$

and the former into the form,

$$1/E = 1/26\,(9/E_{[100]} + 17/E_{[111]}), \qquad \cdots\cdots\cdots\cdots(9)$$

where the term of $1/E_{[110]}$ can be omitted because of the following relation,

$$1/E_{[100]} + 3/E_{[111]} = 4/E_{[110]}\,\cdot \qquad \cdots\cdots\cdots\cdots(10)$$

The relations between Young's modulus of polycrystal calculated by Eqs. (3), (4), (7) and copper contents are shown in Fig. 5. The full lines show Young's modulus of alloys with the grain size which showed the same moduluts as that of nickel. The grain sizes in (a), (b) and (c) are 0.96, 130 and $1000 \times 10^{-4}$ mm$^2$, respectively. As shown in the figure, the theoretical and exerimental curves are considerably deviated from each other. This may be due to the fact that Young's modulus of polycrystal is not determined only by the anisotropy of orientation. Finally, considering that the tendncies of E vs. copper concentration curves are different with different grain sizes, it is desirable to take grain size into account in the relation between a polycrystal and a single crystal.

## Summary

(1) Young's modulus and the grain size have been measured with polycrystalline ferromagnetic nickel-copper alloys annealed at various temperatures.
(2) In the case of heating at constant temperature the grain size is different with different copper concentrations, and the relation between them is different with different heating temperatures.
(3) Young's modulus vs. copper concentration curves by various investigators are inagreeable with one another, and it is considered that the grain size is one of the important causes.
(4) The theoretical equations of Voigt, Reuss and the present authors were compared with one another by using Young's modulus of the single crystals and the experimental results of the polycrystals.

## Appendix

Derivation of the Eq. (7) of the present authors

A few directions having the considerable variation of characteristic and the simple indices were suitably selected, and made themselves responsible for the characteristic of directions near them, and the directions were taken discontinuously. Taking a suitable co-ordinate (XYZ) in single crystal, denote the numbers that the above selected directions apear repeatedly over all directions by

$$k_1,\ k_2,\ k_3,\ \cdots\cdots\cdots\ k_i\ ,\ \cdots\cdots\cdots\ k_j\ .$$

When the crystallites are distributed at random, the probability that the every representative direction agrees with the one selected in the polycrystal becomes

$$k_1/\sigma,\ k_2/\sigma,\ k_3/\sigma\ ,\ \cdots\cdots\cdots\ k_i/\sigma\ ,\ \cdots\cdots\cdots\ k_j/\sigma\ ,$$

where

$$\sigma = k_1 + k_2 + k_3 + \cdots\cdots\cdots + k_i + \cdots\cdots\cdots + k_j\ .$$

The average length of a crystallite is some multiple of dimension ratio $d_i$ and the probability that this appears in a special direction is $k_i/\sigma$, and so the probable value of length becomes

$$l = \sum_{i=1}^{j} d_i\ k_i/\sigma. \qquad \cdots\cdots\cdots\cdots(1)$$

Therefore, the probability that the characteristic of [i]-th direction contributes to this special direction becomes

$$p_i = \frac{d_i\ (k_i/\sigma)}{l}\ . \qquad \cdots\cdots\cdots\cdots(2)$$

Then, the unit that can represent thoroughly the characteristic of polycrystal is

$$\sigma \times l = L. \qquad \cdots\cdots\cdots\cdots(3)$$

Therefore, the total sum of length of [i]-th crystal in this unit is

$$l_i = d_i \times k_i \qquad \cdots\cdots\cdots\cdots(4)$$

Now, denoting Young's modulus of polycrystal specimen by $E$, and that of $L$ direction of crystallite by $E_i$, and assuming that the strain $\delta L/L$ springs up when the stress $T$ is applied from the outside, the following will be obtained:

$$E = \frac{T}{\delta L/L}\ . \qquad \cdots\cdots\cdots\cdots(5)$$

On the other hand, the dimensional variation of [i]-th crystal having $l_i$ in length being $\delta l_i$,

$$\frac{\delta l_i}{l_i} = \frac{T}{E_i}\ . \qquad \cdots\cdots\cdots\cdots(6)$$

The dimensional variation $\varDelta L$ of the above-mentioned unit becomes

$$\varDelta L = \sum_{i=1}^{j}\left(\frac{\delta l_i}{l_i}\right) \times l_i = \sum_{i=1}^{j}\left(\frac{T}{E_i}\right) \times d_i\, k_i\ . \qquad \cdots\cdots\cdots\cdots(7)$$

Moreover, the length $L$ of unit is

$$L = \sigma \sum_{i=1}^{j} d_i\ \frac{k_i}{\sigma} \qquad \cdots\cdots\cdots\cdots(8)$$

Therefore,

$$\frac{\delta L}{L} = \sum_{i=1}^{j}\ \frac{T}{E_i} \times p_i\ . \qquad \cdots\cdots\cdots\cdots(9)$$

Young's modulus $E$ of polycrystal is

$$1/E = \sum_{i=1}^{j} \frac{1}{E_i} \times p_i \cdot \qquad \cdots\cdots\cdots\cdots(10)$$

Now, for the cubic crystal,

$$k_{[100]} = k_1 = 6, \qquad k_1/\sigma = 6/26,$$

$$k_{[110]} = k_2 = 12, \qquad k_2/\sigma = 12/26,$$

$$k_{[111]} = k_3 = 8, \qquad k_3/\sigma = 8/26.$$

Assuming that the every crystallites is the sphere of the same size,

$$d_i \equiv 1. \qquad \cdots\cdots\cdots\cdots(11)$$

Therefore, $$l = \sum_{i=1}^{j} d_i \frac{k_i}{\sigma} = 1, \qquad \cdots\cdots\cdots\cdots(12)$$

and $$p_i = k_i/\sigma \cdot \qquad \cdots\cdots\cdots\cdots(13)$$

Therefore, $$1/E = \sum_{i=1}^{j} \frac{1}{E_i} \times k_i/\sigma \cdot$$

That is, $$1/E = \frac{1}{13}\left(\frac{3}{E_{[100]}} + \frac{6}{E_{[110]}} + \frac{4}{E_{[111]}}\right) \cdot \qquad \cdots\cdots\cdots\cdots(14)$$