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## Solid Solubility Effect of Metallic Surface Friction

By

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The temperature dependence of the metallic surface friction was studied experimentally for a number of metal pairs by the improved BOWDEN method. In particular, for metal pairs of limited solubility type the "Solid Solubility Effect" embodied by the formula:

$$\mu_{\theta} = (a_{ut-c} - a_{\theta})/k$$

was found. Furthermore, similar phenomena are examined for metal pairs of simple eutectic, continuous solubility, intermetallic compounds, intermediate phase types and solidified mercury as a component. The results seem to be in favour to the local melting theory of BOWDEN *et al.*

### 1. Introduction.

In 1935, BOWDEN *et al.*<sup>(1)</sup> found the "stick-slip" phenomenon of the friction between a metal ball and a metal plate, and explained it by the so-called "local melting theory" that the stick is caused by the solidification after the momentary melting of the rubbed local surface at the instant when the metal ball begins to slip against the metal plate, and it is the cause of friction forces. We have investigated the temperature effect on friction, expecting that on the supposition of the local melting theory, there should be observed some close relations between the melting characters of metals and the friction, i. e. some influences of the external heating upon the stick-slip.

### 2. Experimental Method.

The experiments were carried out essentially similarly to those

\* Presented in part at the annual Tokyo meeting of The Physical Society of Japan, April 29, 1946, at the Takaoka meeting of The Japan Society of Metals, October 16, 1946, and at the Sapporo meeting of the Hokkaidô Branch of The Physical Society of Japan, May 1, 1948, and published preliminarily in the *Journal of Applied Physics* of Japan, 17 (1948) 21 and 25 (in Japanese) and the *Physical Review*, 75 (1949) 1621.

of BOWDEN *et al.* with only alternation that our spring balance is of horizontally vibrating type while BOWDEN's one moves along a vertical arc and results no uniform loading at each position. The load is varied in 10 steps from 0 to 1 kg-wt by the compression of another small spring, in contrary to BOWDEN's case where the load is only of one value. The linearity relation of friction force *versus* load is ascertained and its tangent gives the friction coefficient  $\mu$  directly. The temperature was varied by small nichrome electric furnace brought under the metal plate to be rubbed, and its surface temperature was measured by means of a Pt-Pt-Rh thermocouple and a mV meter.

### 3. Experimental Results.

#### A. PRELIMINARY EXPERIMENT.

Preliminarily, fusible metals Sn(232°C)\* and Pb(327°C) were taken, examining the metal pairs Steel-Sn and Steel-Pb, as well as the same metal pairs Sn-Sn and Pb-Pb. In all cases  $\mu$  does not change continuously with the temperature  $\theta$  but vanishes suddenly at the lower of the melting points of both metals. This was thought at first sight to turn against our expectation.

#### B. METAL PAIRS OF THE LIMITED SOLUBILITY TYPE.

##### i. Sn (232°C) - Pb (327°C), Fig. 1.

Expecting the discontinuous change at the melting point observed above for the same metal pairs likely for the different metal pairs, Sn-Pb was examined.  $\mu$  vanishes, however, at 182°C, a temperature much lower than either of both melting points, which is just the eutectic temperature of the binary alloy system Sn-Pb. Magnified figures of the rubbed surfaces of the samples exhibit the existence of a liquid state along the rubbed trace on the Pb-plate.

At the first stage, we thought simply that a certain alloy should be formed at the friction boundary by virtue of the so-called local melting and hence  $\mu$  should vanish at the eutectic point which can be regarded as the melting point of the alloy corresponding to the true melting point in the case of the same metal pair, and further-

\* The temperature bracketted in the rear of the name of metal denotes its melting point in °C.

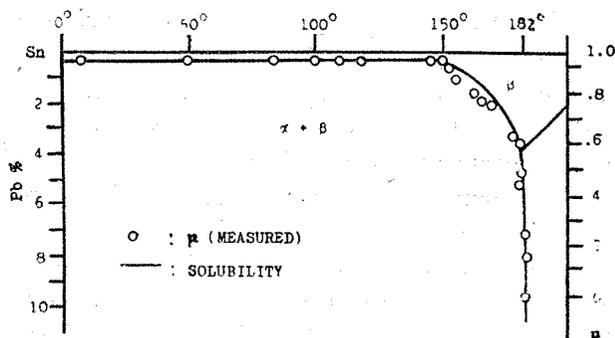


Fig. 1. Friction coefficient  $\mu$  versus temperature  $\theta$  plots brought into phase diagram in Sn-side for Sn-Pb.

more this may be the only temperature effect to be observed. Similar phenomena were also observed for the other different metal pairs, too, so that we stated that it could be called "Eutectic Effect"\* of metallic surface friction.

At the next stage, when not only the point of  $\mu = 0$  but also the full course of  $\mu$  versus  $\theta$  curve ( $\mu$ -curve) were determined precisely, it was found that  $\mu$  begins to decrease gradually at  $146^\circ\text{C}$  already and vanishes finally at  $182^\circ\text{C}$  described above. According to HONDA and ABE<sup>(2)</sup>,  $182^\circ\text{C}$  is a transformation point where Pb begins to solve much more rapidly into Sn-rich  $\beta$ -solid solution.

Then, we noticed that there is such a great similarity between the  $\mu$ -curve and the solid solubility  $\alpha$  versus  $\theta$  curve ( $\alpha$ -curve) in the Sn-side that these two curves can be brought into each other with a simple conversion ratio 10, i. e. 0.1 change of  $\mu$  to 0.01 change of solubility of Pb into Sn. To ascertain the similar phenomena for the other different metal pairs, we examined the following metal pairs of the limited solubility type on referring to JAENECKE's Handbuch der Aller Legierungen<sup>(A)</sup> and HANSEN's Aufbau der Zweistofflegierungen<sup>(B)</sup>.

## ii. Sn ( $232^\circ\text{C}$ ) - Bi ( $271^\circ\text{C}$ ), Fig. 2.

$\mu$  decreases remarkably with temperature rise and vanishes finally at the eutectic point  $139^\circ\text{C}$ , as expected. The  $\mu$ -curve coincides with the  $\alpha$ -curve in the Sn-side of the phase-diagram given by SATÔ and MATSUHASHI<sup>(3)</sup> among many proposed ones of this system,

\* This nomenclature was used in the preliminary report presented at the Tokyo meeting of The Physical Society of Japan, April 29, 1946.

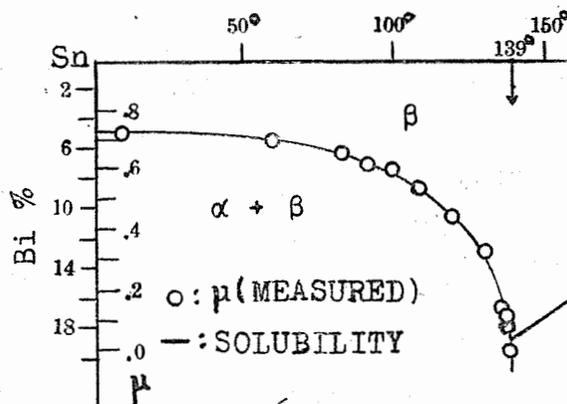


Fig. 2.  $\mu$ - $\theta$  plots brought into phase diagram in Sn-side for Sn-Bi.

assuming 0.1 change of  $\mu$  to correspond to 0.02 change of solubility of Bi into Sn, i. e. conversion ratio 5, in contrary to 10 in the preceding case Sn-Pb.

iii. Sn (232°C)-Cd (321°C), Fig. 3.

The  $\mu$ -curve shows a kink at about 130°C and  $\mu$  vanishes at 182°C. It coincides with the  $\alpha$ -curve given by MATSUYAMA<sup>(4)</sup> very well with the conversion ratio 10. According to him, 182°C is the eutectic point and 130°C is a transformation point of eutectoid.\*

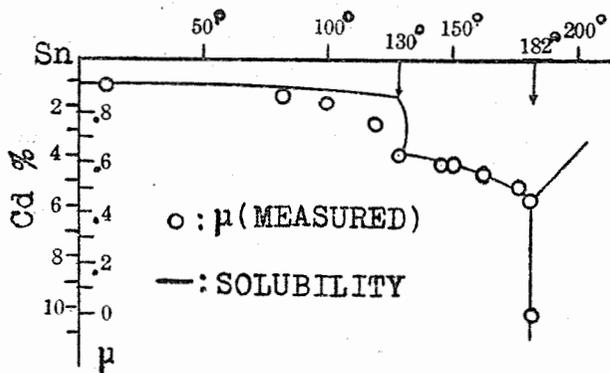


Fig. 3.  $\mu$ - $\theta$  plots brought into phase diagram in Sn-side for Sn-Cd.

iv. Pb (327°C)-Sb (630°C), Fig. 4.

The phase diagrams of this system are slightly different from

\* JAENECKE Handbuch gives a smooth solubility curve in the Sn-side and the eutectic point at 177°C.

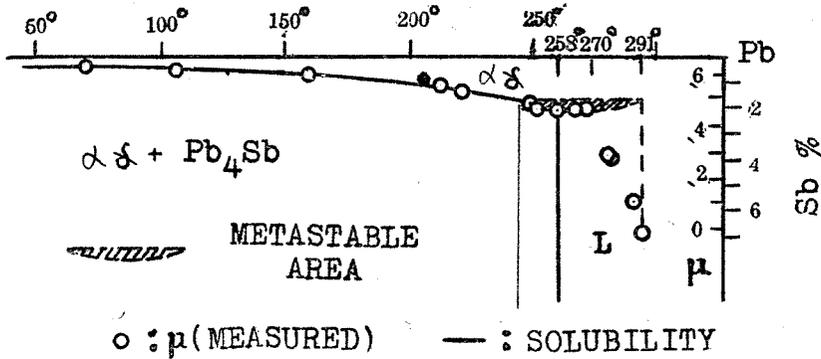


Fig. 4.  $\mu$ - $\theta$  plots brought into phase diagram in Pb-side for Pb-Sb.

each other among many researchers, among which some state even no solubility in Pb-side. The  $\mu$ -curve coincides with the conversion ratio 10 to the  $\alpha$ -curve given by SCHUMACHER and BOUTON<sup>(5)</sup> showing a small solubility in the Pb-side by means of thermal analysis. In this case  $\mu$  does not vanish at the eutectic point 258°C but at 291°C. This can be explained as follows. In the vicinity of the eutectic point there are two solidus curves almost parallel to the temperature axis (drawn in Fig. 4 by broken and chain lines) which indicate the existence of a sort of metastable state (hatched domain in Fig. 4) so that on passing over the eutectic point and on reaching at the kink point 291°C of the chain lined solidus curve they begin to fuse out. On the other hand, the broken lined solidus curve lets well understand the circumstances that the  $\mu$ -curve begins to fall suddenly at about 270°C and the premelting is observed on the rubbed trace at about 278°C, the kink point of the broken lined solidus curve. Thus the  $\mu$ -curve gives not only the general feature of the solubility curve but also some informations about the existence of the metastable state as well as such small solubility as undetectable by usual methods.

v. Bi (271°C)-Pb (327°C), Fig. 5.

The  $\mu$ -curve of this system descends at first in a steep inclination and shows at about 80°C a sudden kink and then runs along the eutectic line gently and finally ends with  $\mu = 0$  at the point of solubility limit. The phase diagram of this system is not uniquely determined among researchers. The kink at 80°C of our  $\mu$ -curve corresponds to that of the solubility curve given by SOLOMON and

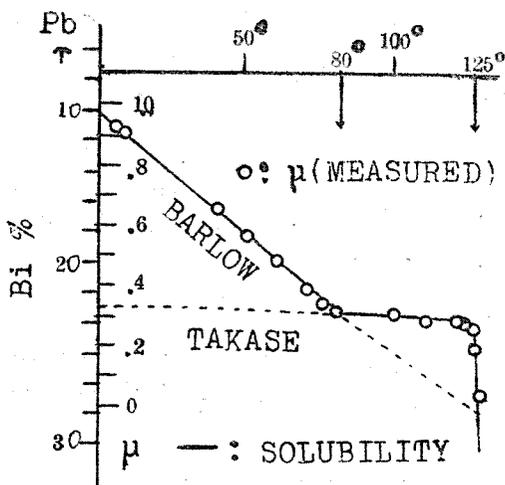


Fig. 5.  $\mu$ - $\theta$  plots brought into phase diagram in Pb-side for Pb-Bi.

metal Pb, in particular, below 80°C with the BARLOW's and above 80°C with the TAKASE's.  $\mu = 0$  corresponds to the solubility limit  $\alpha = 0.3$  at the eutectic point. Consequently, the decrease 1 of  $\mu$  corresponds to the increase 0.2 of  $\alpha$ , so that the conversion ratio is likely 5 as in the similar case Sn-Bi where Bi is also concerned.

vi. Cd (321°C)-Zn (419°C), Fig. 6.

The  $\mu$ -curve coincides well with the  $\alpha$ -curve in the side of the lower melting point metal Cd in the phase-diagram given by JENKINS<sup>(9)</sup> with the conversion ratio 10.

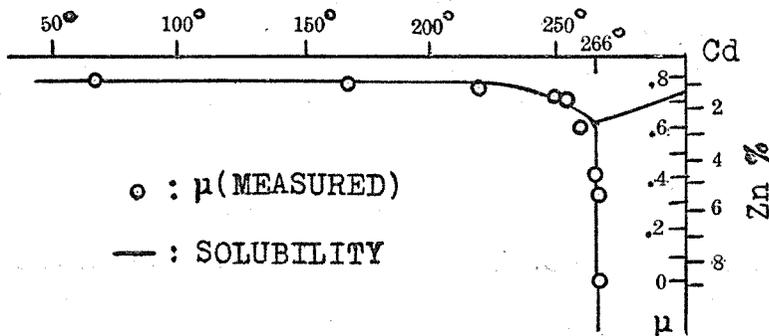


Fig. 6.  $\mu$ - $\theta$  plots brought into phase diagram in Cd-side for Cd-Zn.

vii. Summary, TABLE I.

As seen from TABLE I, many other metal pairs show likely such

JONES<sup>(6)</sup> X-ray-graphically likewise at 80°C, as well as to the cross point of the solubility curve given by BARLOW<sup>(7)</sup> and that given by TAKASE<sup>(8)</sup> each other. Taking  $\mu = 1$  at 0°C (extrapolated) to correspond to  $\alpha = 0.1$  of Bi at 0°C given by BARLOW, and placing the 80°C kinks of both curves upon another, the  $\mu$ -curve coincides with the  $\alpha$ -curve not in the side of the lower melting point metal Bi, but in the side of the higher melting point

similar phenomena that  $\mu$  vanishes generally at the eutectic point and the  $\mu$ -curve can be brought into the solid solubility curve with an appropriate conversion factor, that is embodied by the formula

$$\mu_{\theta} = (\alpha_{\text{eutec}} - \alpha_{\theta})/k, \quad (1)$$

where  $\alpha_{\theta}$  and  $\alpha_{\text{eutec}}$  are the solubilities at the temperature  $\theta$  and the eutectic point respectively, and  $k$  is the conversion factor. The formula can be generalized in such a way as

$$\mu_{\theta} - \mu_{\theta_0} = (\alpha_{\theta_0} - \alpha_{\theta})/k, \quad (2)$$

where  $\theta_0$  is a certain temperature chosen as a standard. Thus we wish to call this phenomenon as "Solid Solubility Effect" of metallic surface friction.

TABLE I. *Temperature of vanishing friction coefficient and value of factor  $k$  in Eq. (1) for several metal pairs of limited solubility.*

Metal pair	Lower melt. pt.	Eutec. pt.	Temp. $\mu = 0$	$k$ for weight %	$k$ for atomic %
Sn—Pb	232°C	182°C	182°C	10	5.9
Sn—Bi	232	139	139	20	12
Sn—Cd	232	182	182	10	10.4
Sn—Zn	232	199	199	10	10
Sn—Tl	232	170	170	10	10
Bi—Pb	271	125	125	20	20
Bi—Cd	271	148	148	*	*
Bi—Zn	271	254	254	*	*
Tl—Cd	302	204	204	10	10
Cd—Pb	321	247	247	10	10
Cd—Zn	221	255	255	10	16.5
Pb—Sb	327	258	291**	10	16.5

\* Solubility is too small to determine the value of  $k$ .

\*\* This coincides with the kink point of the solidus line.

**Character 1.** The conversion factor  $k$  takes the different value according to whether the solubility  $\alpha$  is expressed in atomic concentration or in weight one, and in the weight percentage  $k$  takes noticeably a constant value 10 for every metal pairs except when Bi is concerned, while in the atomic percentage it is not the case. (TABLE I)

**Character 2.** The solubility curve must be taken in the side of the

lower melting point metal, namely in the side where the atoms of the higher melting point metal dissolve into the lower melting point metal.

**Character 3.** The only exception is the case where Bi is concerned. In this case,  $k$  has a value 5 and the solubility curve must be taken in the side where Bi dissolve into the other partner metal irrespectively of its melting point. This fact is quite interesting when we recall the statement of HOFE and HANEMANN<sup>(10)</sup> that the mobility of Bi in the partner metal lattice is greater than that of the latter in Bi.

**Theoretical consideration.** The linearity relation between  $\mu$  and  $\alpha$  can be explained by the fact that the condition for minimum free energy of the rubbed boundary layer determines at each temperature the most stable state giving the solubility of alloys and the maximum friction coefficient  $\mu$  which measures the degree of resistance to the mobility of atoms. The value 10 of  $k$  may be understood as follows. According to BRAUE and BARRER<sup>(11)</sup> the entropy of diffusion of the solute in metallic diffusion process is  $\Delta S_d = 20$  cal/deg per mol at the melting point of the solvent, while the entropy of melting is  $\Delta S_m = 2$  cal/deg per mol, and the ratio of these values gives just the dimensionless numerical value 10. The theoretical consideration along this line is now being studied more precisely.

**Exceptional case 1.** Al-Ag, Al-Zn, Al-Mg, Al-Sn.

In these systems, the solid solubility effect is not observed, whereas they are of the type of limited solubility which should change notably with temperature as before. This is due to the oxyde film of a quite high melting point which is so steady as to prevent even the diffusion of small oxygen atom when the coated film has grown up to some extent, so that in our experiment the partner metal atom is forbidden to diffuse as solute any more through the rubbed boundary layer. Therefore, when the amount of the diffused atoms is not so much, the solid solubility effect cannot be seen in the complete form, as it is the case, for example, for some simple eutectic alloy systems.

**Exceptional case 2:** Sn-Tl, Zn-Bi.

Although  $\mu$  vanishes at the eutectic point,  $\mu$  has not a definite value corresponding to the small solubility, but rather rises with temperature. This seems to be due to the great stick-slip, but its

dependences on the relative translation velocity and the proper frequency of the experimental apparatus are being further investigated.

### C. METAL PAIRS OF SIMPLE EUTECTIC ALLOY TYPE.

In order to see what rôle the existence of eutectic crystal may play, we examined metal pairs of the so-called simple eutectic type which have no mutual solubility (TABLE II).

TABLE II. *Simple eutectic alloy type.*

Metal pair	Sn—Ag	Sn—Al	Pb—Ag	Cd—Bi	Sn—Zn
Melting point	232° 960°	232° 659°	327° 960°	321° 271°	232° 419°
Eutectic point	221°	229°	304°	146°	199°
$\mu = 0$ point	232°	232°	327°	146°	199°

These pairs do not show the expected eutectic effect. In the last two cases in TABLE II, however, if we wait for a sufficiently long time we can see  $\mu$  to vanish at the eutectic point. This means that there is slight solubility between them (2 to 0.2% in Sn—Zn case)<sup>(2)</sup> (Fig. 7). The similar effects should be verified in the first three

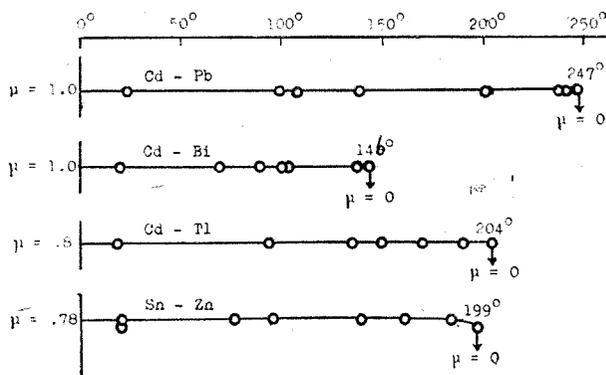


Fig. 7.  $\mu$ - $\theta$  plots for Cd-Pb, Cd-Bi, Cd-Tl and Sn-Zn.

cases, too, but it is very difficult because the eutectic point is near to the melting point of pure metal. From these facts we see that the essential point is not eutectic type of the metal pair but rather mutual solubility between them, so that the terminology of "Solid Solubility Effect" is more appropriate than "Eutectic Effect".

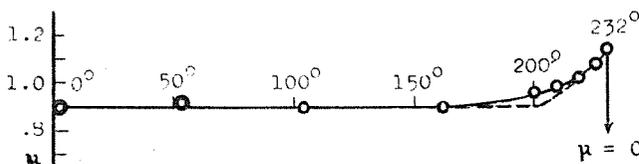
## D. METAL PAIRS OF CONTINUOUS SOLUBILITY TYPE.

The metal pair which is mutually soluble at any ratio, i. e. so-called "lückenlos" soluble, has rather high melting point, so that it is difficult to trace the phenomena up to the melting point. Generally we see, the larger the solubility is, the smaller  $\mu$  is. For example, in the case of Cu (1083°C)-Ni (1455°C),  $\mu$  is very small (about 0.56) and nearly constant up to the measured temperature 400°C.

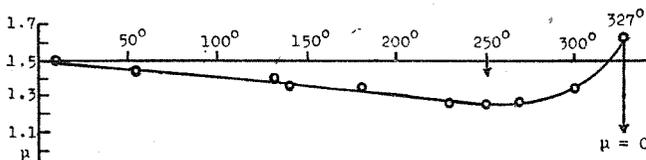
## E. METAL PAIRS OF INTERMETALLIC COMPOUNDS TYPE.

i. Mg(651°C)-Sn(232°C), actually Mg<sub>2</sub>Sn(778°C)-Sn(232°C), Fig. 8.

In this system, Mg<sub>2</sub>Sn and pure Sn form a simple eutectic alloy. The  $\mu$ -curve is a straight line parallel to the temperature axis as expected by virtue of the vanishing solubility between them. At the eutectic point (200°C)  $\mu$  does not vanish, but rather begins to increase and suddenly falls to zero from the maximum value at the melting point of Sn (232°C).

Fig. 8.  $\mu$ - $\theta$  plots for Mg-Sn.ii. Mg(651°C)-Pb(327°C), actually Mg<sub>2</sub>Pb(550°C)-Pb(327°C), Fig. 9.

In this system, Mg<sub>2</sub>Pb and pure Pb have the eutectic point at 250°C. Corresponding to the existence of some solubility in the Pb-side, the  $\mu$ -curve falls linearly with temperature up to 250°C, and then begins to rise and suddenly falls from the maximum value to zero at the melting point of Pb (327°C), similarly to the preceding case Mg-Sn. In these two systems the eutectic point is certainly a singular point, while there happens no melting.

Fig. 9.  $\mu$ - $\theta$  plots for Mg-Pb.

iii. Mg(651°C)-Zn(419°C), actually MgZn<sub>5</sub>(500°C)-Zn(419°C), Fig. 10.

In this system, MgZn<sub>2</sub>(590°C) and pure Zn form a simple eutectic alloy, while the  $\mu$ -curve runs parallel to the temperature axis similarly to the system Mg-Sn,  $\mu$  does not vanish at the eutectic point between MgZn<sub>5</sub> and pure Zn, 364°C, but unexpectedly at the peritectic point between MgZn<sub>2</sub> and MgZn<sub>5</sub>, 381°C.

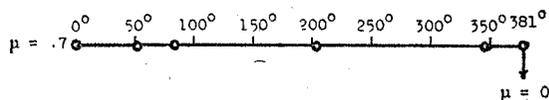


Fig. 10.  $\mu$ - $\theta$  plots for Mg-Zn.

#### F. METAL PAIRS OF INTERMEDIATE PHASE TYPE.

i. In(156°C)-Ag(960°C), Fig. 11.

JAENECKE and HANSEN state that the eutectic point between  $\varphi$ -phase of In-Ag and pure In is 141°C and there is no solubility in the In-side. Our  $\mu$ -curve shows, however, that  $\mu$  vanishes at 139°C. According to WEIBKE and EGGERS<sup>(3)</sup>, the solidification temperature varies slightly with the Ag concentration in In as shown in TABLE III and "eutektikum" is at 2.25% and 140°C. Therefore our result implies that the eutectic point is at about 140°C and the corresponding solubility is of about 1%. At lower temperatures than 90°C there are some fluctuations of  $\mu$  which can be understood from the fact that the lower the temperature is the smaller the solubility is.

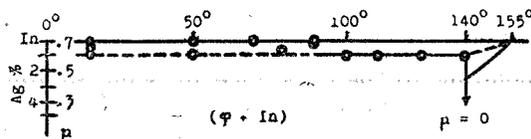


Fig. 11.  $\mu$ - $\theta$  plots brought into phase diagram in In-Side for In-Ag.

TABLE III. Solidification temperature of In-Ag.

Ag weight, %	1	2	3	5
Eutectic point	139°	140°	140°	141°

ii. In(156°C)-Cu(1083°C), Fig. 12.

JAENECKE and HANSEN state that the eutectic point between  $\varphi$ -

phase of In-Cu and pure In is 153°C. Our  $\mu$ -curve shows, however, that  $\mu$  does not vanish at the eutectic point 153°C but at the melting point of In 156°C, whereas at 153°C a tendency of falling is slightly observable. According to WEIBKE and EGGERS<sup>(14)</sup> the solidification temperature is constant independently of the Cu-concentration in In, as shown in TABLE IV.

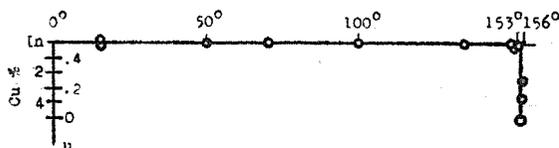


Fig. 12.  $\mu$ - $\theta$  plots brought into phase diagram in In-side for In-Cu.

TABLE IV. Solidification temperature of In-Cu.

Cu weight %	1	2.22	3.3	4.5
Eutectic point	153°	153°	153°	153°

Hence our result enforces that there is probably no solubility in this case, in contrary to the preceding case In-Ag.

iii. Pb-Tl, Ag-Zn, Ag-Cd, Cu-Zn, Sn-Sb, Fig. 13, 14, 15, 16.

The phase diagrams of these binary systems are divided completely into many intermediate phases forming intermetallic compounds. The measured  $\mu$ -curves have a similarity to their phase boundary curves. By virtue of the non-existence of the eutectic point,  $\mu$  vanishes rather at the lower of the melting points of the component metals.

In particular, in the Cu-Zn case (Fig. 16),  $\mu$  has at lower temperatures two different values, according as whether the rubbing ball is of Zn or of Cu, but these two curves converge with rising temper-

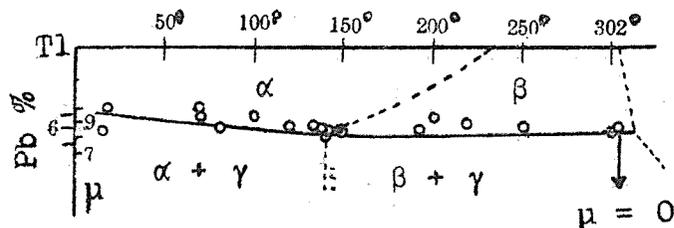
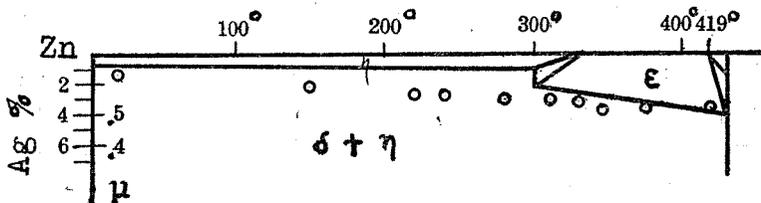
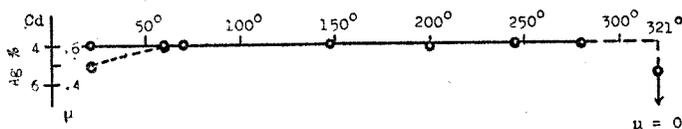
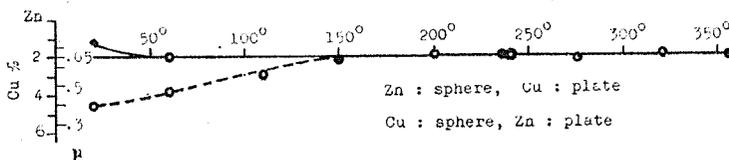


Fig. 13.  $\mu$ - $\theta$  plots brought into phase diagram in Tl-side for Pb-Tl.

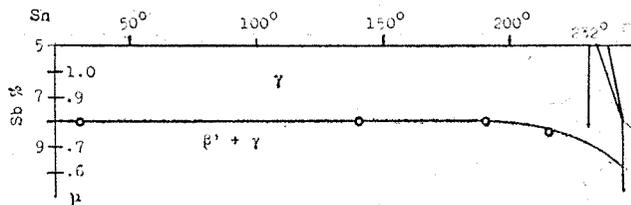
Fig. 14.  $\mu$ - $\theta$  plots brought into phase diagram in Zn-side for Ag-Zn.Fig. 15.  $\mu$ - $\theta$  plots brought into phase diagram in Cd-side for Ag-Cd.Fig. 16.  $\mu$ - $\theta$  plots brought into phase diagram in Zn-side for Cu-Zn.

atures into one line. This may be ascribed to the fact that the contact of the rubbed surface does not occur exactly in one point.

Generally, for the intermetallic compound type, we cannot state decisively a quantitative relation between  $\mu$  and solubility.

#### iv. SnSb ( $\beta'$ phase, stable up to 325°C)—Sn (232°C), Fig. 17.

If there are many intermediate phases it is not simply clear, which of those phase boundaries corresponds really to the  $\mu$ -curve. In order to avoid this ambiguity, we formed really  $\beta'$ -phase of Sn-Sb and measured  $\mu$  between that and pure Sn. The results are similar to the simple Sn-Sb case described above, clarifying the behavior more precisely. According to IWASE<sup>(16)</sup>, the boundary between the

Fig. 17.  $\mu$ - $\theta$  plots brought into phase diagram in Sn-side for Sn-Sb.



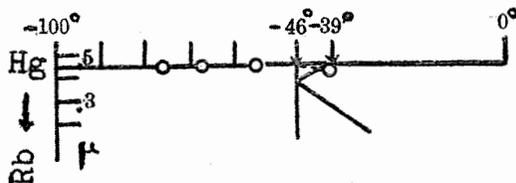


Fig. 19.  $\mu$ - $\theta$  plots brought into phase diagram in Hg-side for Hg-Rb.

iii. Hg ( $-39^{\circ}\text{C}$ )-Te ( $452^{\circ}\text{C}$ ), Fig. 20.

$\mu$  is constant up to the melting point of Hg,  $-39^{\circ}\text{C}$ . We found no singular point on the  $\mu$ -curve. The solubility diagram in the Hg-side is not yet known while PELLINI<sup>(17)</sup> gives only the Te-side. From our  $\mu$ -measurement we suppose no eutectic point to exist in the Hg-side in favour to the expected diagram given in HANSEN'S book (the broken line in Abb. 322).

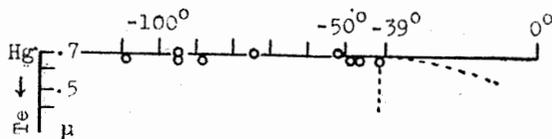


Fig. 20.  $\mu$ - $\theta$  plots brought into phase diagram in Hg-side for Hg-Te.

From these experiments we see that there exist some diffusion velocity even at much lower temperatures than the melting point of Hg, resulting the solid solubility effect and suggesting the existence of the slight solubility or the exact form of the phase diagram.

#### 4. Conclusion.

We have found some characteristic relations between friction coefficient  $\mu$  and temperature  $\theta$  for many different metal pairs, in particular, for metal pairs of limited solubility type the "Solid Solubility Effect" embodied by the formula:

$$\mu_{\theta} = (\alpha_{\text{eutec}} - \alpha_{\theta})/k.$$

Furthermore, similar phenomena are examined for metal pairs of simple eutectic, continuous solubility, intermetallic compounds and intermediate phase types. These phenomena can be reversely used to check the phase diagram or to construct it.

All experiments described above seem to be in favour to the local melting theory of BOWDEN *et al.*

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