EFFECT OF SOLUTE CONCENTRATION ON WORK HARDENING BY COLD ROLLING

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ABSTRACT

Accumulative reciprocal rolling and one pass rolling of letter "U" type specimen were applied to pure aluminum, Al-Zr, Al-Cu, Al-Mg and Al-1.0%Si solid solutions. Excepting the solid solutions showing obvious effects of G.P. zone formation or precipitation, separation of contribution to resistivity of dislocation increased by the rolling from that of vacancy was attempted through room temperature holding for 86.4ks to 15Ms. Higher solute concentration leads to a larger increase in resistivity due to dislocation for a same rolling true strain. Al-Mg solid solutions up to 8.10% were rolled to various true strain and showed work hardening and resistivity increase due to dislocation both of which are larger in higher concentration of Mg.

Keywords: *resistivity, dislocation density, rolling effect, solute effect, concentration effect*

1. BACKGROUND, MOTIVATION AND AIM.

Starting from Matthiessen's rule, Komatsu et al. [1, 2] induced a relation between resistivity ratio $R=\rho_{300}/\rho_{77}$ and ρ_{77}. Under correction with deviation from Matthiessen's rule (DMR), empirical equations expressed as $\rho_{77}=\alpha/(R-\eta)+\beta$ have been obtained for many solid solution systems, where α and β are constants and η represents the ratio of contribution to resistivity per unit concentration of solute atoms $\Delta\rho$ at 300K and 77K, $\Delta\rho_{300}/\Delta\rho_{77}$. In binary solid solution, α can be written as $\rho^{PURE}{}_{300}-\eta\rho^{PURE}{}_{77}$, where ρ^{PURE} is the resistivity of ideally pure solvent metal at temperature of suffix. However in ternary or more complicated real solutions, the best way is to determine the α and β empirically by annealing the specimen of which precise size factor can be measured.

Utilizing these DMR corrected empirical equation (DMRCEE), one can estimate the ρ_{77} from the R of specimens difficult to measure the size factor. This method has been already applied to 1050 aluminum alloys and has detected Si precipitation during holding at room temperature (RTH) for 86.4ks, 1 day after cold rolling [3].

In a series of studies on dependence of resistivity of Al-Mg alloys on solute Mg concentration [4] and annealing, Komatsu et al. [5] observed that the work hardening by 92% cold rolling showed a parabolic increase with Mg content whereas the increment in resistivity showed a linear increase. Moreover, the relation between the resistivity increment and the amount of work hardening, or difference in proof stress of the 92% cold rolled state from the 623K-3.6ks annealed state, fitted well to the relation, $\delta\sigma_{0.2}/\mathrm{MPa}=263(\delta\rho/\mathrm{n\Omega m})^{1/2}$, where $\delta\sigma_{0.2}=\sigma_{0.2}{}^{CR}-\sigma_{0.2}{}^{ANN}$ and $\delta\rho=\rho_{77}{}^{CR}-\rho_{77}{}^{ANN}$ [5].

Hirsch and Mitchell [6] proposed the relation between dislocation density and work hardening as $\delta\sigma=\alpha Gb(N^{DISL})^{1/2}$, where α is a non-dimensional constant from 0.5 to 1.0, G is the rigidity modulus, b is the Burgers vector and N^{DISL} is the dislocation density. Writing the contribution to resistivity per unit density of dislocation as $\Delta\rho^{DISL}$ one can obtain $N^{DISL}=\delta\rho^{DISL}/\Delta\rho^{DISL}$, and $\delta\sigma=[\alpha Gb(\Delta\rho^{DISL})^{-1/2}]\bullet[(\delta\rho^{DISL})^{1/2}]$. Employing $\Delta\rho^{DISL}=3\times10^{-25}\ \Omega\mathrm{m}^3$ [7], α for Al-0 to 4%Mg was estimated as 0.59 [5], which seems to be a reasonable value. This fact suggests that the dislocation density increased by cold rolling well corresponds to the resistivity increase after RTH for more than 10-30Ms.

In above case of the fixed reduction of thickness to 92%, the tendency, larger work hardening in higher solute concentration, was obvious. The next investigation should aim to whether this tendency appears in other solid solution than Al-Mg and for Al-Mg alloys whether the above tendency appears in various rolling reduction.

2. EXPERIMENTAL PROCEDURE

1.0mm or 0.5mm^t × 10mm^W × 150mm^L plates obtained by cold rolling were formed within nitride steel mold to letter "U" type by sawing and filing as schematically illustrated in **Fig. 1**. Solute content and constants in the DMRCEE are shown in **Table 1** with heat treatment to determine the constants and ρ_{M77} after annealing or reversion before the rolling. The rolled specimen was immersed into a liquid nitrogen bath immediately after leaving the roll, within 1 s. The total time from start of the rolling till the immersing into liquid nitrogen was less than 15s, in the hand driven reciprocal rolling.

Because the resistivity very quickly decreases at room temperature immediately after the rolling, the resistance at 300K, Ω_{300}, is firstly measured after 86.4ks (1 day) holding at room temperature. Until the room temperature holding (RTH) for 86.4 ks, only Ω_{77} was measured to follow the change by the RTH. We call the resistivity at 77K calculated from the DMRCEE and measured $R=\Omega_{300}/\Omega_{77}$ as ρ_{M77} to distinguish from the measured resistivity using density method size factor, ρ_{p77}. Other details of the procedure to obtain Matthiessen size factor M.F. and ρ_{M77} immediately after the rolling [3] are shown in **Fig. 2**. Accumulative reciprocal rolling (ARR) and repeated RTH give only a rough information about vacancy concentration and dislocation density induced by rolling. No rolling of spot-welded four terminal leads is the important point of this letter "U" type specimen method. Therefore, one way and one pass rolling is possible for the "U" type specimen having prerolled four fingers end [8]. Same procedure to obtain change in the ρ_{M77} as the ARR can be applied also to the one pass rolling.

Al-Mg specimens rolled and shaped to 0.5×3×80mm were measured the ρ_{M77} after the RTH for more than 11 Ms, then tensile tested at room temperature with strain rate of 6.7×10^{-4} s^{-1}. Because specimens have no chucking part, or have uniform cross sectional area for full length, this tensile test does not fit to any industrial standard. However, in our experience, the value of proof stress is not affected largely by this form of specimen.

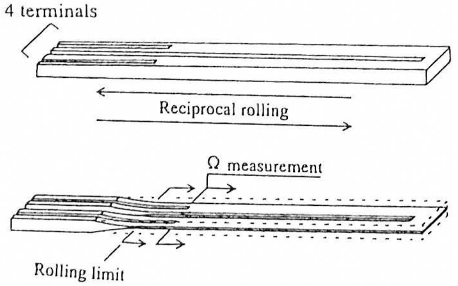

Fig. 1. Schematic illustration of letter "U" type specimen before and after accumulative reciprocal rolling.

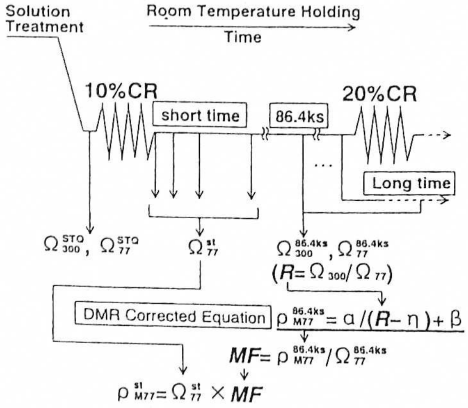

Fig. 2. Procedure to estimate the resistivity immediately after rolling and its change during room temperature holding (RTH).

Table 1. Solute content, constants in the DMR corrected empirical equation $\rho_{77}=\alpha/(R-\eta)+\beta$, heat treatment to determine the constants and ρ_{M77} after annealing or reversion before the rolling.

Constant of DMR corrected equation ;

$\rho_{77}/\mathrm{n\Omega m}=\alpha/(R-\eta)+\beta$

	mass%	α	β	η	Treatment
pure Al	0.0001	25.13	–	1	〈Theoretical〉
	0.001	25.13	–	1	
	0.01	25.61	−0.038	1	Long period Ic.T.(50K/864ks) 〈CR~423K and Full Annealing〉
Mg	0.47	28.42	+0.648	0.88	
	0.93	27.87	+0.478		
	1.45	28.92	−0.644		
	1.89	30.27	−1.202		
	2.87	31.07	−0.678		
	3.79	30.60	+0.160		
	5.05	31.07	−0.678		
Zr	0	25.13	–	1	〈Theoretical〉
	0.029	25.43	−0.095	1.2	Long period Ic.T. (50K/7.2ks) 〈CR~473K〉
	0.059	23.07	+0.183		
	0.086	25.15	−0.133		
	0.117	20.53	+0.728		
	0.153	23.20	+0.204		
Cu	0.46	23.83	+0.138	1	Ic.T.(50K/3.6ks) 〈CR~373K and Full Annealing〉
	1.01	24.81	+0.038		
	1.52	26.33	−0.318		
	1.95	24.00	+0.279		
	3.02	22.34	+0.878		
	4.04	20.19	+2.222		
Si	1.0	24.28	+0.282	1.06	Ic.T.(50K/3.6ks),〈CR~423K, F.A.〉

3. WORKING HYPOTHESES AND RESTRICTIONS.

Fig. 3 shows a result of the ARR applied to an Al-Zr specimen. RTH for 86.4ks looks enough to saturate the decrease in resistivity which is assumed to correspond with annihilation of point defects, perhaps mainly vacancy, produced by the cold rolling. In other words, total increase in ρ within 1 s after the rolling can be divided into two parts, $\delta\rho^{TOTAL}=\delta\rho^{DISL}+\delta\rho^{VAC.}$, as illustrated schematically in right hand side of the Fig. 3. The residual increase after the 86.4ks RTH should be an overestimation of maximum 30%, which one can read in left hand side of the Fig. 3 from decrease by the RTH for 14.1Ms after 90% ARR.

There are many exceptions of this assumption. **Fig. 4** shows the change in ρ_{M77} of Al-3.02%Cu and -4.04%Cu alloys during long period RTH after the one pass rolling. The G.P. zone formation seems to occur even during the rolling within a few seconds and continues through the RTH. The ρ_{M77} increases, in contrast to the decrease in Fig. 3, for 10 and 30% rolling. Another exception is Al-1.0% Si alloy in **Fig. 5**. Because the equilibrium solubility of Si to Al at 300K is extrapolated as 3.5~6.0×10^{-6}mass%, even in a 1050 aluminum alloy containing only 0.09%Si the decrease in ρ_{M77} by the RTH exceeded the increase by cold rolling [3]. This excess decrease in ρ appears more obviously in Al-1.0%Si solid solution and one can observe in Fig. 5 that the ρ_{M77} becomes lower than the value before rolling from 12.8% cold reduction. For the Al-3.79%Mg alloy, the ρ_{M77} increases by the RTH for 180s up to 22% of the ARR, as shown in **Fig. 6**.

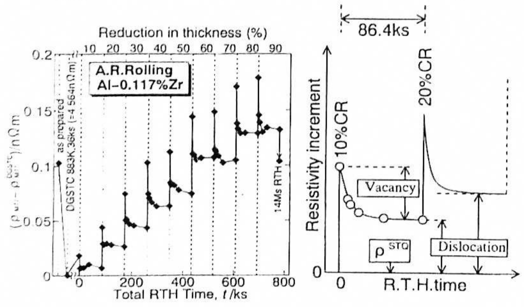

Fig. 3. Change in ρ_{M77} of Al-0.117%Zr with the accumulative reciprocal rolling and during room temperature holding for 86.4ks (left) and schematic illustration of model for dividing the contribution of dislocation and vacancy (right).

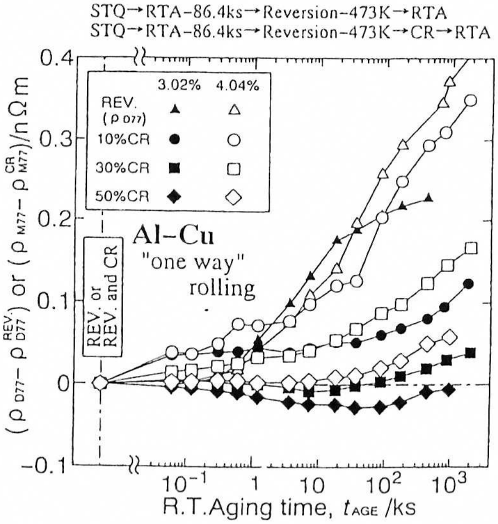

Fig. 4. Changes in ρ_{M77} of Al-3.02%Cu and -4.04%Cu alloys by room temperature aging after reversion for 0.3ks at 473K and one pass rolling of various reduction.

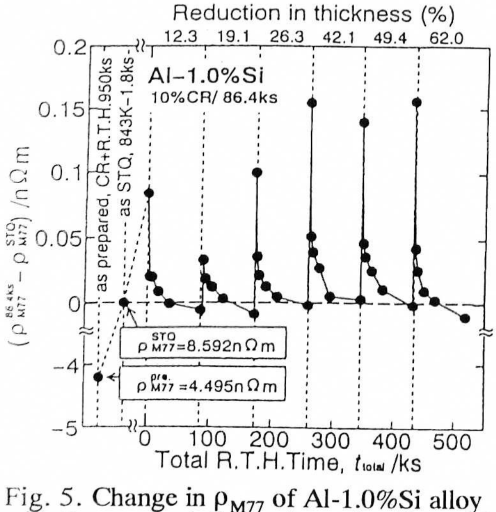

Fig. 5. Change in ρ_{M77} of Al-1.0%Si alloy with the accumulative reciprocal rolling and during room temperature holding for 86.4ks. No practical increase remains after the RTH.

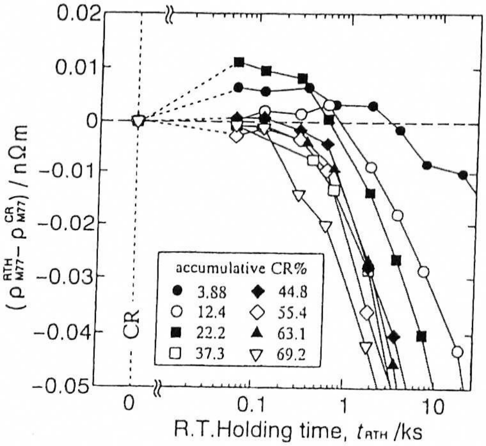

Fig. 6. Change in ρ_{M77} of Al-3.79%Mg alloy by room temperature aging after the ARR of various reduction.

Though the maximum increment is not so large, only 0.01nΩm in 3.79%Mg alloy and even in 8.10% Mg alloy that was 0.05nΩm, corresponding 1 to 2% of total increment respectively, the ARR of "U" type specimen was applied to only alloys containing below 1.45%Mg, below 1.95%Cu, Al-Zr alloys and 4N, 5N and 6N pure aluminum in present work. The one pass rolling was applied to more concentrated Al-Mg and Al-Cu alloys.

4. RESULTS AND DISCUSSION.

4.1 EFFECT OF SOLUTE CONCENTRATION ON RESISTIVITY INCREASE BY ACCUMULATIVE RECIPROCAL ROLLING.

Fig. 7 shows relations between rolling true strain, ε, and resistivity increment in pure aluminum by the ARR. The resistivity increases parabolically with true strain. For ε above 1.0, the increment by rolling is obviously larger in less pure specimens. A long period RTH slightly lowered the residual increase in the ρ_{M77} of 6N-Al.

Fig. 8 compares the decrement by the 86.4ks RTH to the residual increment after the RTH as a function of ε. Both changes in ρ_{M77} are parabolic to the ε and are large in high concentration solid solution. **Fig. 9** shows log-log plots for the $\delta\rho^{DISL}$ and ε. In dilute solutions, the plot gives fairly good linearity. **Table 2** shows constants A and n in regression according to $\delta\rho/n\Omega m = A\varepsilon^{n}$ for both $\delta\rho^{DISL}$ and $\delta\rho^{VAC.}$. The exponent n for $\delta\rho^{DISL}$ varies from 0.4 to 0.8, however, the constant A clearly increases with solute concentration.

Using constants in Table 2, the $\delta\rho^{DISL}$ at 50% reduction (ε=0.693) is calculated for various systems and compared in **Fig.10** as function of solute concentration in at.%, except pure aluminum specimens of which purity is given only in mass%. It is clear that the $\delta\rho^{DISL}$ at a same rolling strain increases with solute concentration. Plots of each group give good straight lines which can be written as $\log(\delta\rho^{DISL}) = \log\lambda + \mu\log(C/\text{at\%}, \text{ or mass\%})$. **Table 3** shows constants λ and μ for four groups. The exponent μ is larger in Al-Cu and Al-Mg systems than in Al-Zr and pure aluminum systems. The whole plots in Fig. 10 seem to make a broad band having shape of inverse L. $\delta\rho^{DISL}$ by the ARR was larger than that by one pass rolling in more concentrated alloys, e.g. Al-3.02%Cu and Al-3.79%Mg alloy, because of effect of G.P. zone formed during the repeated RTH. Value of $\delta\rho^{DISL}$ after about 50% one pass rolling and RTH for 0.86Ms was plotted also in the Fig. 10. Perhaps because of log scale, the coincidence to the results of ARR is fairly good.

4.2 WORK HARDENING AND RESISTIVITY INCREMENT.

Though alloys containing more than 1.89%Mg showed increase or stagnation of decrease in resistivity, the alloys up to Al-8.10%Mg were rolled aiming to 50, 75 and 83% reduction getting 0.5mm as final thickness.

After RTH for 11Ms the rolled specimens were finally measured the resistivity and tensile tested. Increment in proof stress from the annealed state at same ε showed a tendency to increase with the Mg content. However, the increment of $\delta\rho^{DISL}$ was rather scattered with the Mg content, because too short specimens, only 80mm compared to 180mm for ρ_{D77} measurement, were used.

Results are shown in **Fig.11** as the relation between $(\delta\rho^{DISL})^{1/2}$ and $\delta\sigma_{0.2}$. The solid line in the Fig. 11 is previous result obtained from 92% cold rolled plates containing up to 3.79%Mg [5]. There is a tendency that the plots for higher concentration alloys give larger work hardening and resistivity increase due to dislocation, just same as result for 92% cold rolling. Moreover, alloys containing more than 5.05%Mg deviate lower side of the previous result up to 3.79%, which can be explained by the overestimation of $\delta\rho^{DISL}$ through G.P. zone formation during the RTH or somewhat short period (11 Ms) of present RTH.

5. CONCLUSIONS.

Though repeated the accumulative reciprocal rolling and room temperature holding for 86.4ks, 20 to 30% overestimate the resistivity increase due to dislocation density increased by rolling, it is suggested that the higher concentration of impurities or solute atoms leads to higher dislocation density at a same rolling strain.

Evidence of G.P. zone formation during room temperature holding or aging after rolling has been obtained. In the Al-3.78%Mg alloy rolled 50%, the maximum increment of resistivity is only 0.01nΩm which corresponds only 1% of total increment by rolling. In Al-4.04%Cu alloy, the resistivity continues to increase until for longer 1 Ms aging at room temperature up to the increment of 0.35nΩm.

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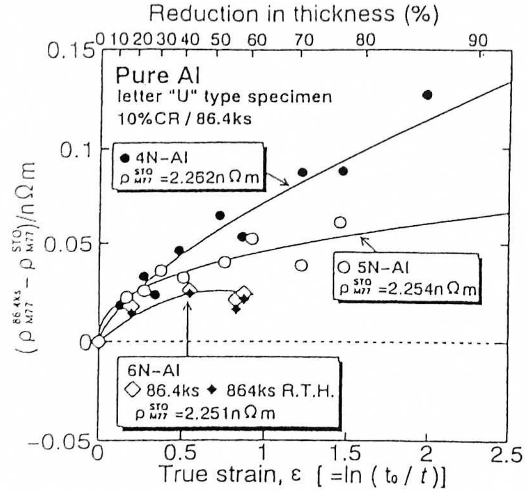

Fig. 7. Relations between true rolling strain and resistivity increment due to dislocation by the ARR and RTH for 86.4ks for pure aluminum of various purity.

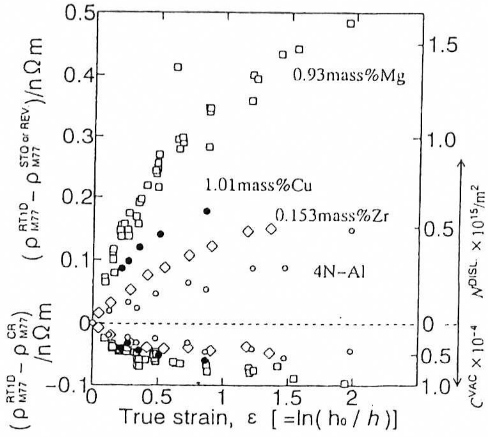

Fig. 8. Comparison of the decrement during 1 day RTH with the residual increment after the RTH, $\delta\rho^{DISL}$, as function of true rolling strain ε.

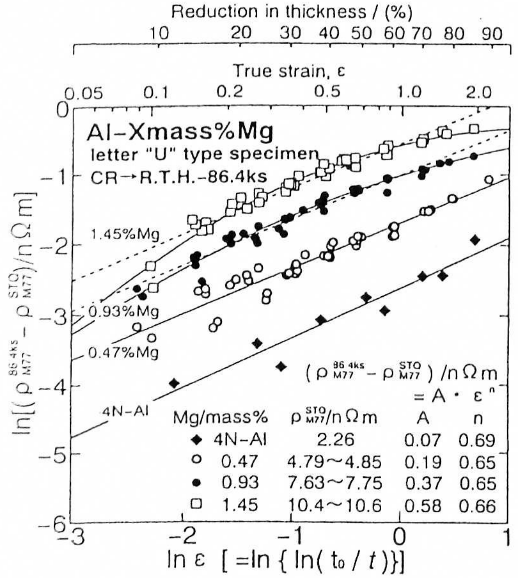

Fig. 9. Log-log plots for the $\delta\rho^{DISL}$ and ε, according to the relation $\delta\rho^{DISL} = A\varepsilon^n$.

Table 2. Constants A and n in $\delta\rho/\text{n}\Omega\text{m} = A\varepsilon^n$ for $\delta\rho^{DISL}$ and $\delta\rho^{VAC.}$.

solute / mass%		$\rho^{DISL.} = \rho^{RT1D} - \rho^{STO}$		$\rho^{VAC.} = \rho^{CR} - \rho^{RT1D}$	
		A/nΩm	n	A/nΩm	n
	6N-Al	0.027	0.174	–	–
	5N-Al	0.046	0.396	0.041	0.610
	4N-Al	0.073	0.716	0.040	0.358
Mg	0.47	0.185	0.648	0.048	0.272
	0.93	0.367	0.646	0.072	0.379
	1.45	0.579	0.656	0.093	0.361
Zr	0	0.075	0.631	0.040	0.419
	0.029	0.078	0.717	0.028	0.608
	0.059	(0.071)	(1.109)	0.037	0.521
	0.086	0.094	0.592	0.044	0.407
	0.117	0.119	0.797	0.045	0.382
	0.153	0.127	0.687	0.046	0.484
Cu	0.46	0.124	0.589	0.053	0.397
	1.01	0.202	0.521	0.063	0.376
	1.52	0.318	0.456	0.087	0.618
	1.95	0.417	0.408	0.100	0.640

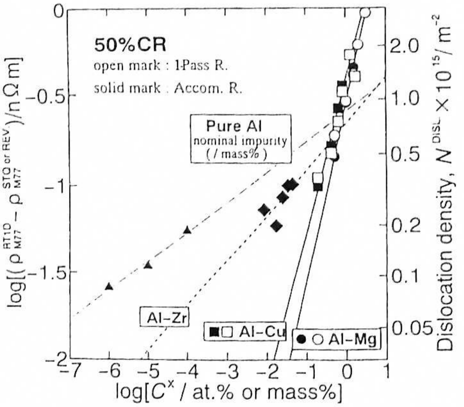

Fig.10. Relation between impurity concentration/mass% or solute concentration/at% and calculated $\delta\rho^{DISL}$ at 50% reduction in thickness using constants in Table 2.

Table 3. Constants λ and μ in $\log(\delta\rho^{DISL})=\log\lambda+\mu\log(C/\text{at\%}$, or mass%) for four solid solution groups.

Constants in regression :

$$\delta\rho^{DISL}/n\Omega m=\lambda\,(C/\text{at\% or mass\%})^{\mu}$$

	λ	μ	r
pure Al	0.258	0.169	0.990
Al-Zr	0.221	0.258	0.738
Al-Cu	0.392	0.871	0.988
Al-Mg	0.282	1.011	0.999

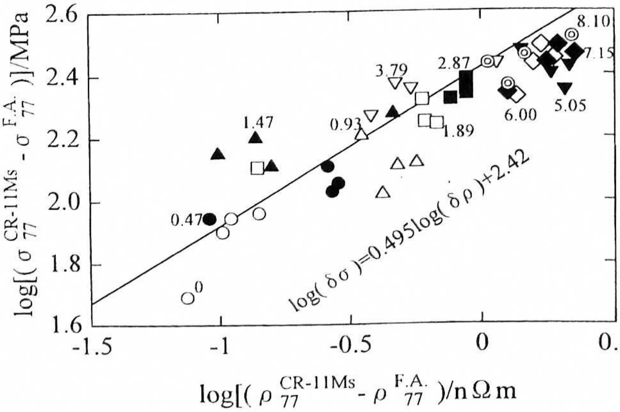

Fig. 11. Relation between $(\delta\rho^{DISL})^{1/2}$ and $\delta\sigma_{0.2}$ for Al-Mg alloys rolled to 50, 75, 83 and 92%.

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