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ELECTRICAL RESISTANCE OF ALUMINUM AT LOW TEMPERATURES

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ABSTRACT

The electrical resistance of high-purity, commercially available aluminum was measured in the temperature range 2.2 to 30° K. The highest values of resistance ratio $R_{300}/R_{4.2}$ were obtained on samples annealed for 100 hours at 400° C and slowly cooled to room temperature; for zone-refined material the resistance ratio was 6630; for material with a nominal impurity content of 10 ppm the resistance ratio was 2250.

The intrinsic resistivity is not defined uniquely by the temperature but is also a function of the resistance ratio, that is, it is a function of the concentration of lattice defects.

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Introduction

The purpose of this work was to obtain data on the low-temperature electrical resistance of commercially available high-purity aluminum. The data were to be of sufficient accuracy to allow proper design of cryogenic magnets; the literature data are not sufficiently complete to allow for proper design.

The resistance as a function of temperature has been measured by Boorse and Niewodniczanski,[1] Caron[2] and Powell et al.[3], as well as many other earlier investigators; however, an examination of the data disclosed that neither the Bloch-Gruneisen relation nor Matthiesen's rule is followed exactly; that is, specimens of different purity give different values of intrinsic resistivity at the same temperature. Further, the data of Wernick et al.[4] and Albert et al.[5] indicated that commercially available high-purity aluminum gives a resistance ratio of $R_{273}/R_{4.2}$ of 1500 and zone-refined material gives resistance ratios in the range of 5000 to 7000; it was then of interest to measure the intrinsic resistivity of this type of material as a function of temperature. Since the above resistance ratios had been obtained on massive specimens, it was also of interest to ascertain whether or not appreciable impurity pickup takes place during fabrication of the ingots into more useful shapes.

Experimental

The objectives of the experimental program were to determine (1) the optimum annealing conditions by measuring the resistance ratio $R_{300}/R_{4.2}$ as a function of annealing conditions, and (2) the temperature dependence of resistance of samples annealed under different conditions.

The materials used were the Super-Raffinal and Zone-Refined grades of aluminum manufactured by Aluminum-Industrie-Aktien-Gesellschaft, which contain the impurities listed in Table 1.

Table 1. Impuritics in Aluminum
(Nominal impurities as given by the manufacturer)

	Super-Raffinal 99.999% pure	Zone-Refined 99.9999% pure
Si	1.5 ppm	not detected
Fe	2.0 ppm	0.3 ppm
Cu	0.8 ppm	0.4 ppm
Ti	not detected	not detected
Mg	not detected	< 0.2 ppm
Zn	not detected	not detected
Ba	---	not detected
Co	---	not detected

All the annealing experiments were carried out using specimens made from 24-gauge, Super-Raffinal aluminum wire. At first the wire was wound as a 4-terminal resistor on a glass rod, Figure 1, but this was found to be unsatisfactory because it was very easy to strain the wire after annealing and before carrying out the resistance measurements. A new set of wire specimens was made by winding a helix of wire on a mica cross (Figure 1), as is done for the construction of strain-free platinum resistance thermometers. The data obtained using the latter type of specimens were more reproducible. The specimens used in the annealing experiments were made small enough to fit through the neck of a helium transport dewar, and were completely covered by liquid helium during the low-temperature measurements.

The temperature dependence of the resistance was measured on the wire specimens as well as on specimens of 1-mm square cross section, machined from ingots of Super-Raffinal and Zone-Refined aluminum. These machined specimens were carefully cleaned and degreased prior to annealing, and were mounted between two aluminum plates with glass cloth and quartz wool for insulation. Figure 2 shows machined specimens in their holders.

The resistance measurements were made using a Rubicon six-dial thermofree potentiometer and a galvanometer amplifier. To limit heating effects, the measuring current was kept to about 0.9 amp for the wire and 3 amp for the machined samples. The current was measured by the voltage drop across calibrated shunts.

Metallographic examination after the resistance measurements showed that all the samples were polycrystalline.

A cryostat was designed to allow resistance measurements in the temperature range 2 - 30° K. Temperatures from 2.2 to 4.2 and 14 to 20° K were obtained by pumping on liquid helium and liquid hydrogen; temperatures between 4.2 and 14 and above 20° K were obtained by allowing the sample holder to warm up slowly.

The sample holder (machined from electrolytic tough pitch copper) was made in two parts. The lower part was permanently mounted inside the cryostat and contained the carbon resistors and thermocouples used for temperature measurement, as well as the reservoirs for the vapor pressure thermometers. The reservoir used for hydrogen contained a small amount of ortho-para conversion catalyst.

The aluminum samples were securely mounted on the upper part of the same holder, and all the lead-in wires were clamped securely to the sample holder prior to connecting them to the aluminum resistances. Since a number of resistances were measured in the same run, the current flow was arranged in series using relatively heavy copper wires for interconnecting the resistors. These copper wires were also clamped firmly onto the sample holder.

The upper part of the sample holder was lowered into the cryostat, where it rested on a cup containing mercury located on the top of the lower part of the sample holder. The mercury layer provided a large area of good thermal contact between the upper and lower parts of the sample holder. Amalgam formation was slow because the sample holder was not degreased and was held at room temperature or below.

The lower part of the sample holder had a chimney extension, a 1/8-in.-thick-wall copper tube, which extended above the top of the upper part of the sample holder and acted as a radiation shield between the aluminum samples and the walls of the dewar.

In the liquid ranges of hydrogen and helium, the temperatures were measured by the respective vapor pressures and the aluminum resistances were completely immersed in the liquid. Between liquid helium and liquid hydrogen, as well as above 20°K, the temperatures were calculated from the resistance of a carbon resistor which was calibrated for each run and its resistance fitted with an equation of the type

$$1/T = A/\ln R + B + C \ln R + D \ln^2 R + E \ln^3 R \qquad (1)$$

Since the above equation did not give a perfect fit to the calibration points, temperatures between 4.2 and 14.0 are not known absolutely to better than about 0.1°K; the relative values are known to about 0.01°K. The absolute error in temperatures above 20°K is harder to estimate; in that region some readings were also made with a gold-cobalt vs copper thermocouple. Between 4.2 and 14 and above 20°K, the aluminum resistances were cooled by convection by the gas and by conduction along the lead wires.

Annealing Results

The optimum annealing conditions are: about 100 hr at 400°C; slow cooling to room temperature; followed by one week storage at room temperature. The wire specimens annealed in this manner gave resistance ratios $R_{300}/R_{4.2}$ of about 2000, while the machined specimens gave values of about 2200 for Super-Raffinal and 6630 for the Zone-Refined material.

It is not known whether this 10% difference in the resistance ratio between wire and machined specimens of the same nominal purity is due to a size effect or to a small amount of impurity pickup.

In the annealing experiments the following parameters were investigated:

1. Effect of annealing temperature. It was investigated for a 2 hour anneal in air, letting the sample cool down with the furnace — about 1 hour to room temperature. The results are shown in Figure 3; the resistance ratio increases from about 270 for the un-annealed samples to about 1860 for the samples annealed at 500°C. The anomalously low values at 550°C

were due to contamination; the brass screws and washers used to hold the aluminum wire in place in the specimens wound around a glass rod (Figure 1) had welded to the aluminum by diffusion.

2. Effect of quenching. It was found that quenching the samples had, as expected, an appreciable effect on the residual resistance. The resistance ratio for the quenched samples (Figure 4) was about 5% lower than that of the slow-cooled samples. Quenching was obtained by removing the samples from the furnace and letting them air cool to room temperature in a few minutes.

3. Effect of extending cooling schedules. The following cooling schedule from 400°C did not have any appreciable effect on the resistance ratio: 2 hr at 290°C; 16.5 hr at 170°C; and 10.7 hr at 114°C. However, it was found that letting the samples stand at room temperature for about one week increased the resistance ratio; thus, the resistance ratio of a sample increased from 1930 to 1956 in one week but did not change thereafter.

4. Effect of vacuum vs air anneal. There was no appreciable effect on the residual resistance when samples were annealed in a vacuum of about 5 microns of mercury.

5. Effect of annealing time. The resistance ratio increases with longer annealing times (Figure 4) but it seems doubtful that times in excess of 100 hours would do very much good. From Figure 4 it can be noticed that while at long times there is no difference in annealing at 400 or 500°C, the samples annealed at 300°C had appreciably lower resistance ratios.

The resistance measurements on the strain-free wire samples showed a high degree of reproducibility, thus, for a sample of resistance ratio of 1950, successive measurements were reproducible within 0.4%, even though the sample had been disconnected and reconnected to the measuring leads.

Temperature-Dependence Measurements

Measurements of the temperature dependence were carried out on wire samples of resistance ratios $R_{300}/R_{4.2}$ ranging from 276 to 2012 and machined samples of resistance ratios of about 2200 and 6600.

Before examining the data, it is worth while to describe the way in which the corrected sample temperature was obtained. Even though a serious attempt had been made at making the samples isothermal with the sample holder and the temperature sensors, it was realized that some means had to be established to ascertain how good the thermal contact really was, and to obtain the effective temperature of each sample.

To determine this, the data were analyzed as follows: first, using the values of resistance below 4.2 °K, the residual resistance R^0 was obtained by extrapolating to 0 °K.

The values of the intrinsic resistance $R-R^0/R_{300}-R^0$ were then calculated for each resistor and empirically fitted versus temperature using an equation of the form:

$$\ln r = A/T + B + CT + DT^2 \qquad (2)$$

$$r = R-R^0/R_{300}-R^0 \qquad (3)$$

When the deviation of the experimental values from those calculated from equation (2) was plotted versus temperature (Figure 5), it was observed that for some of the resistors there existed a discontinuity in the values of the deviation at about 14 - 15 °K and at 20 °K. Each of the points in Figure 5 corresponds to a potentiometer reading; the points appear in pairs at about the same temperature, corresponding to the two readings obtained upon reversal of the measuring current.

The discontinuity in the values of the deviations can be interpreted readily if it is remembered that the resistance readings from 20° down to about 14°K were obtained with the sample immersed in liquid hydrogen, while the readings from 4.2 up to about 15°K, as well as the readings above 20°K, were obtained while the samples were being cooled by convection by the gas. The discontinuity is then due to the fact that when the samples were being cooled by the gas, their temperature was somewhat higher than the gas temperature. From equation (2) one can obtain the temperature coefficient of resistance, and therefore the extent of the corresponding temperature discontinuity. For the sample shown in Figure 5, this temperature gap was about 0.2°K; for other samples the temperature gap ranged from 0 to 0.5°K, and one sample that had a loose electrical connection gave a temperature gap of about 1.5°K.

From the value of this temperature gap and estimated values of the heat transfer coefficient from the aluminum resistors to the gas and to the liquid, a set of temperature corrections can be obtained and applied to all the measurements. When the corrected values of temperature were fitted using equation (2), the deviations between the observed and calculated values no longer showed a gap (Figure 6).

In Figures 7 and 8 the reduced resistance is plotted versus the corrected temperature. The data in the low-temperature region (Figure 7) are plotted vs T^5. While the scatter in the data is such that it is not possible to say whether the relation between r and T^5 is linear or not below 4.2°K, it is clear that appreciable deviations from linearity occur for values of T^5 of about 3800 ($T = 5.2$°K).

A very interesting result is presented in Figure 8, which gives values of the reduced resistance in the temperature region 6 to 22°K. Each sample has a different temperature coefficient of resistance; if the values of r at a given temperature are plotted versus the resistance ratio, a smooth curve is obtained for values of $R_{300}/R_{4.2}$ ranging from 1834 to 2270. For this reason the data obtained on samples of resistance ratios 1921 and 1979 are not shown in Figure 8. It should be remembered that these aluminum samples are of the same nominal purity and differ only in the degree of anneal; the wire samples were all obtained from the same spool. The results are listed in Table 2.

It is unfortunate that the temperature coefficient was not measured on samples of resistance ratios between 276 and 1834 and between 2200 and 6600.

Table 2. Reduced Resistance, $R-R^{\circ}/R_{300}-R^{\circ}$ versus Temperature for Aluminum Samples

T	Resistance ratio $R_{300}/R_{4.2}$							
(°K)	6630	2270	2242	2012	1979	1921	1834	276
2	$1. \times 10^{-7}$							
3	$8. \times 10^{-7}$							
4	2.7×10^{-6}							
5	$6. \times 10^{-6}$							
6	1.13×10^{-5}							
7	1.70×10^{-5}	1.80×10^{-5}						1.82×10^{-5}
8	2.46×10^{-5}	2.78×10^{-5}						2.94×10^{-5}
9	3.32×10^{-5}	4.50×10^{-5}	4.13×10^{-5}					4.52×10^{-5}
10	4.33×10^{-5}	6.40×10^{-5}	5.75×10^{-5}	5.90×10^{-5}				6.68×10^{-5}
11	5.51×10^{-5}	8.2×10^{-5}	7.52×10^{-5}	7.98×10^{-5}				9.40×10^{-5}
12	6.84×10^{-5}	1.02×10^{-4}	9.56×10^{-5}	1.02×10^{-4}			1.06×10^{-4}	1.24×10^{-4}
13	8.38×10^{-5}	1.25×10^{-4}	1.18×10^{-4}	1.27×10^{-4}			1.34×10^{-4}	1.57×10^{-4}
14	1.01×10^{-4}	1.50×10^{-4}	1.44×10^{-4}	1.57×10^{-4}	1.63×10^{-4}	1.60×10^{-4}	1.65×10^{-4}	1.96×10^{-4}
15	1.19×10^{-4}	1.77×10^{-4}	1.73×10^{-4}	1.89×10^{-4}	1.95×10^{-4}	1.91×10^{-4}	1.99×10^{-4}	2.42×10^{-4}
16	1.43×10^{-4}	2.06×10^{-4}	2.05×10^{-4}	2.26×10^{-4}	2.32×10^{-4}	2.30×10^{-4}	2.37×10^{-4}	2.92×10^{-4}
17	1.69×10^{-4}	2.46×10^{-4}	2.43×10^{-4}	2.67×10^{-4}	2.73×10^{-4}	2.72×10^{-4}	2.78×10^{-4}	3.53×10^{-4}
18	2.01×10^{-4}	2.90×10^{-4}	2.84×10^{-4}	3.13×10^{-4}	3.20×10^{-4}	3.21×10^{-4}	3.28×10^{-4}	4.31×10^{-4}
19	2.39×10^{-4}	3.40×10^{-4}	3.36×10^{-4}	3.66×10^{-4}	3.76×10^{-4}	3.74×10^{-4}	3.84×10^{-4}	5.20×10^{-4}
20	2.86×10^{-4}	3.96×10^{-4}	3.94×10^{-4}	4.23×10^{-4}	4.35×10^{-4}	4.36×10^{-4}	4.52×10^{-4}	6.30×10^{-4}
21				4.57×10^{-4}			5.30×10^{-4}	7.22×10^{-4}
22				5.77×10^{-4}			6.25×10^{-4}	8.38×10^{-4}
23								9.50×10^{-4}
24								1.08×10^{-3}
25								1.22×10^{-3}
26								1.37×10^{-3}

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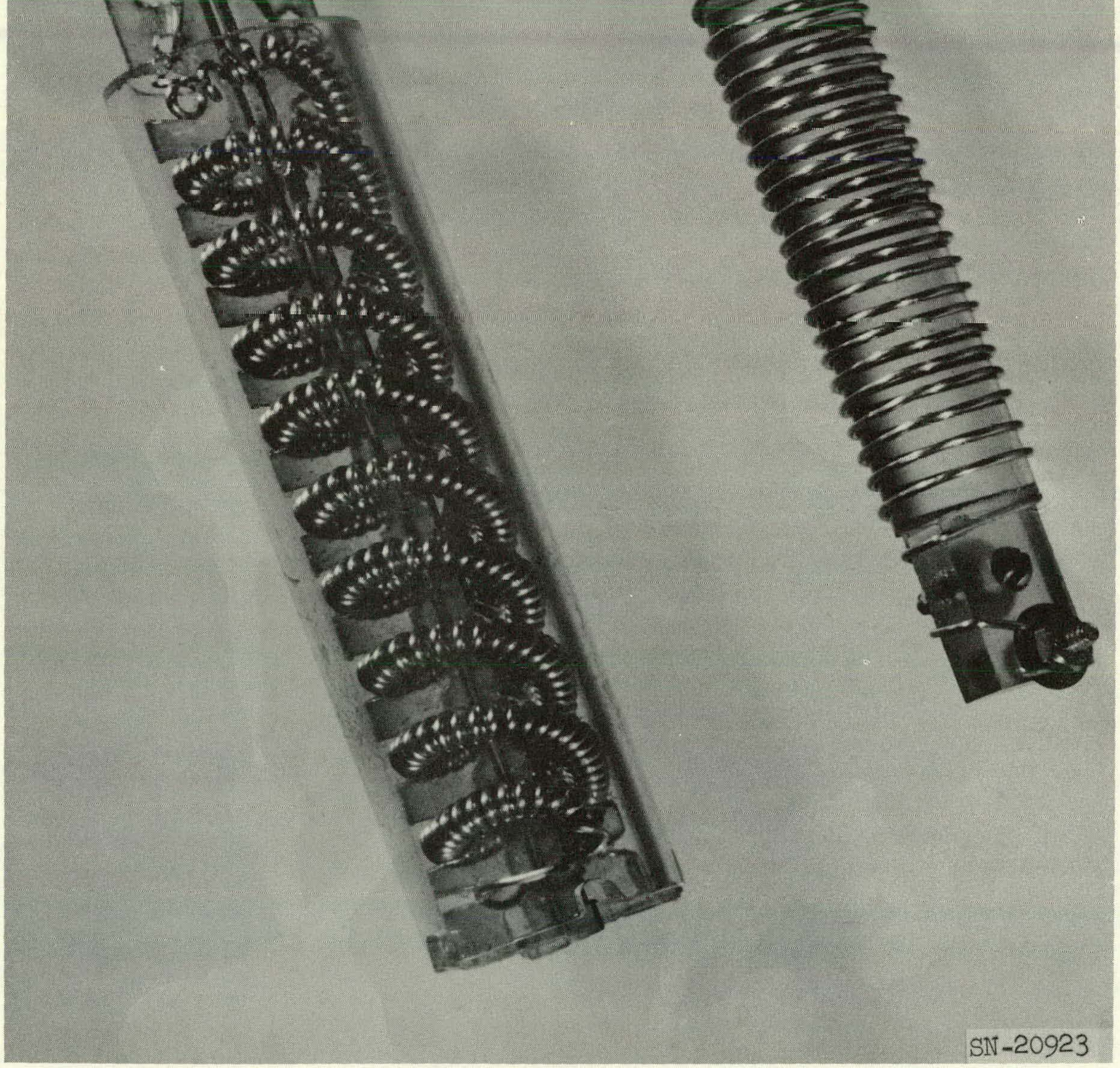

Figure 1. Aluminum wire specimens

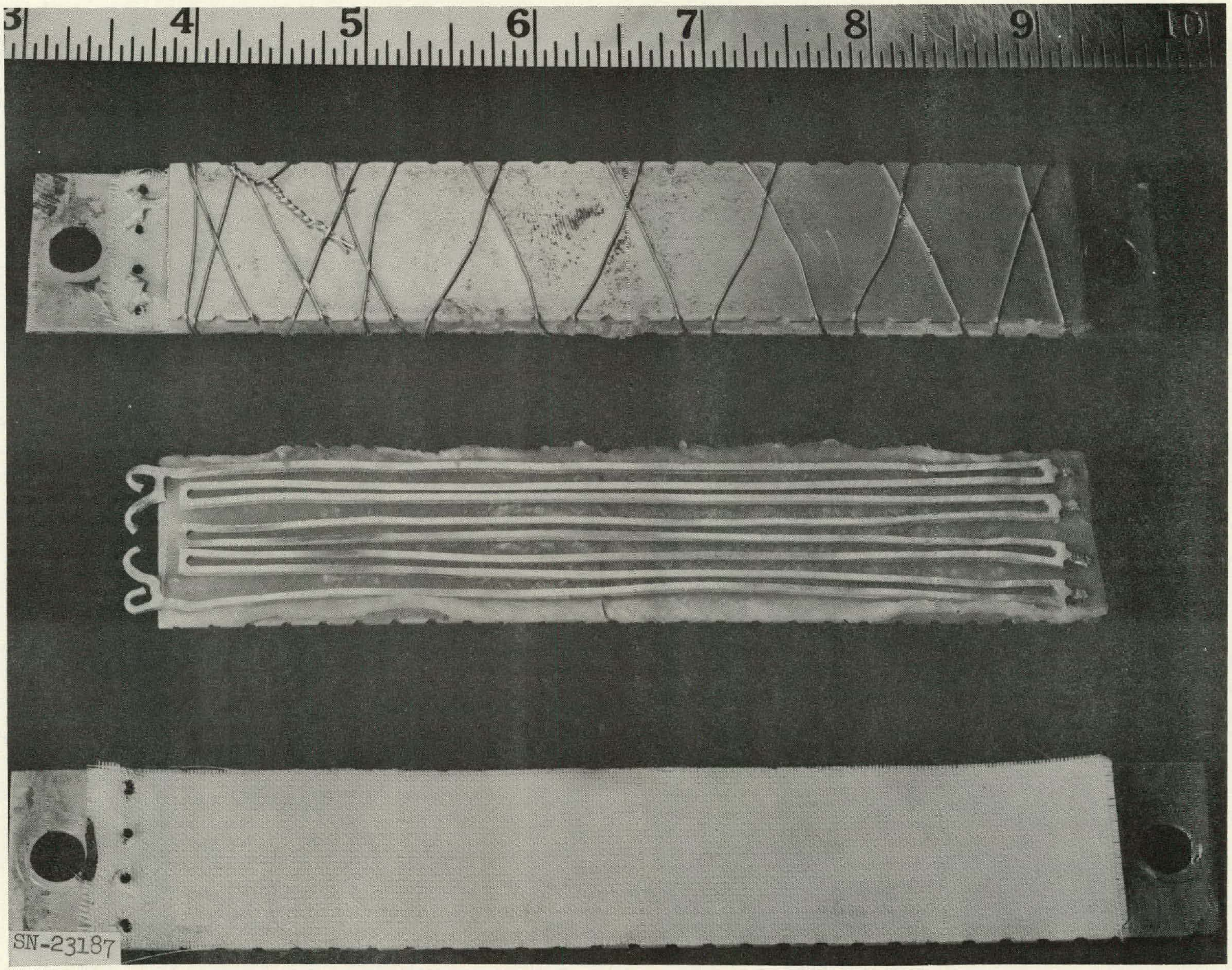

Figure 2. Aluminum machined specimens

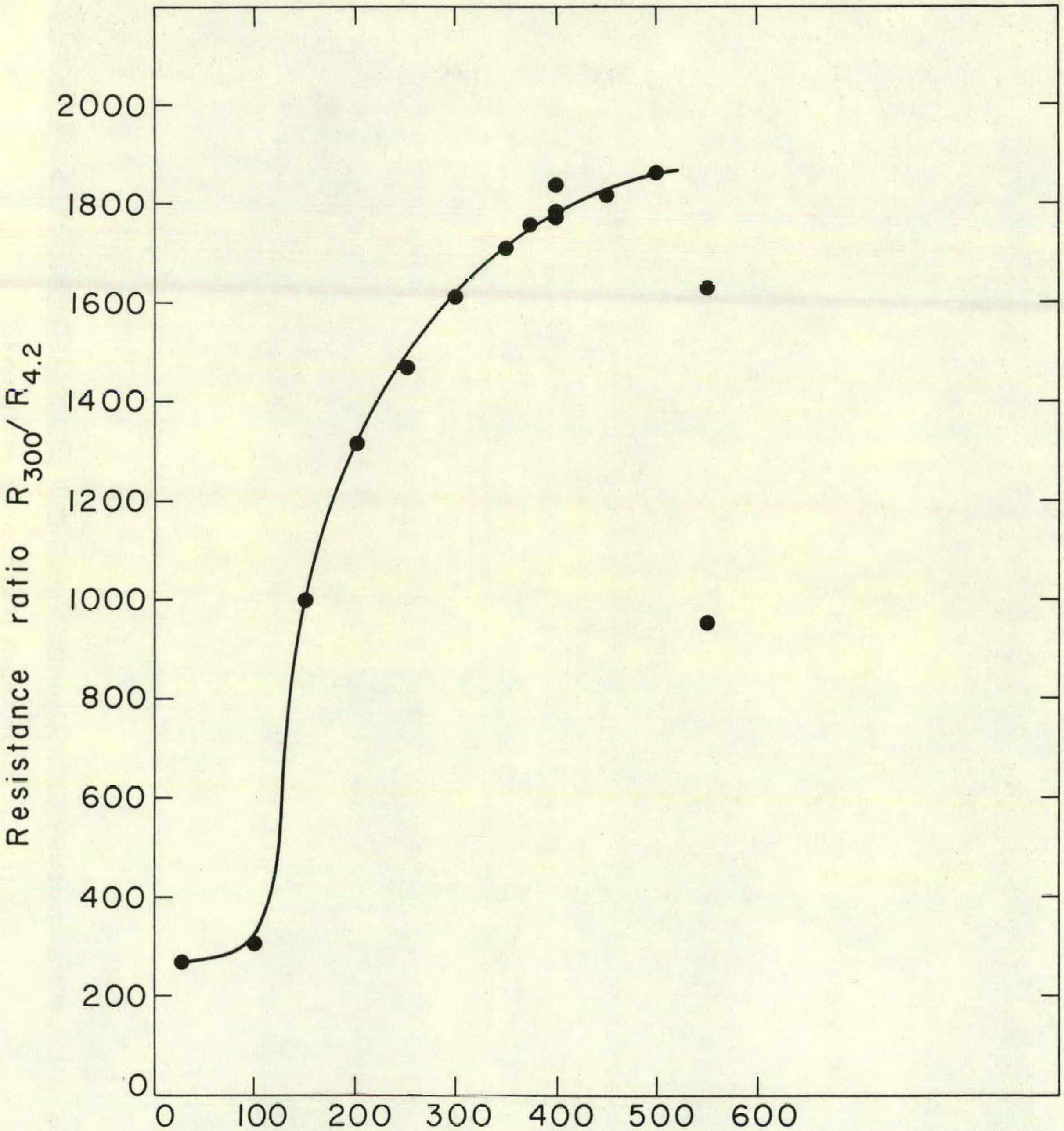

Figure 3. Resistance ratio vs annealing temperature for a 2-hour anneal

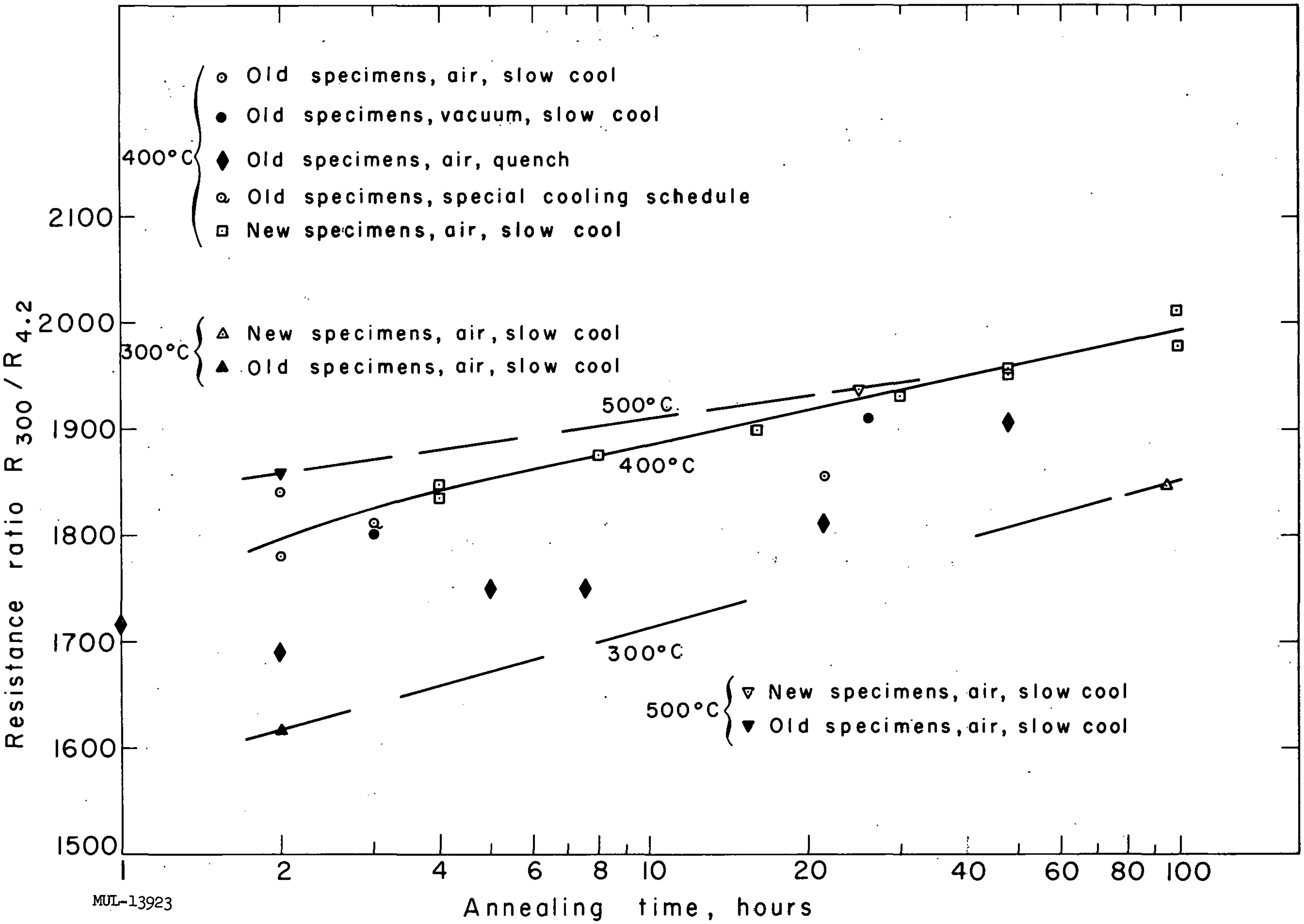

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Figure 4. Resistance ratio vs annealing time

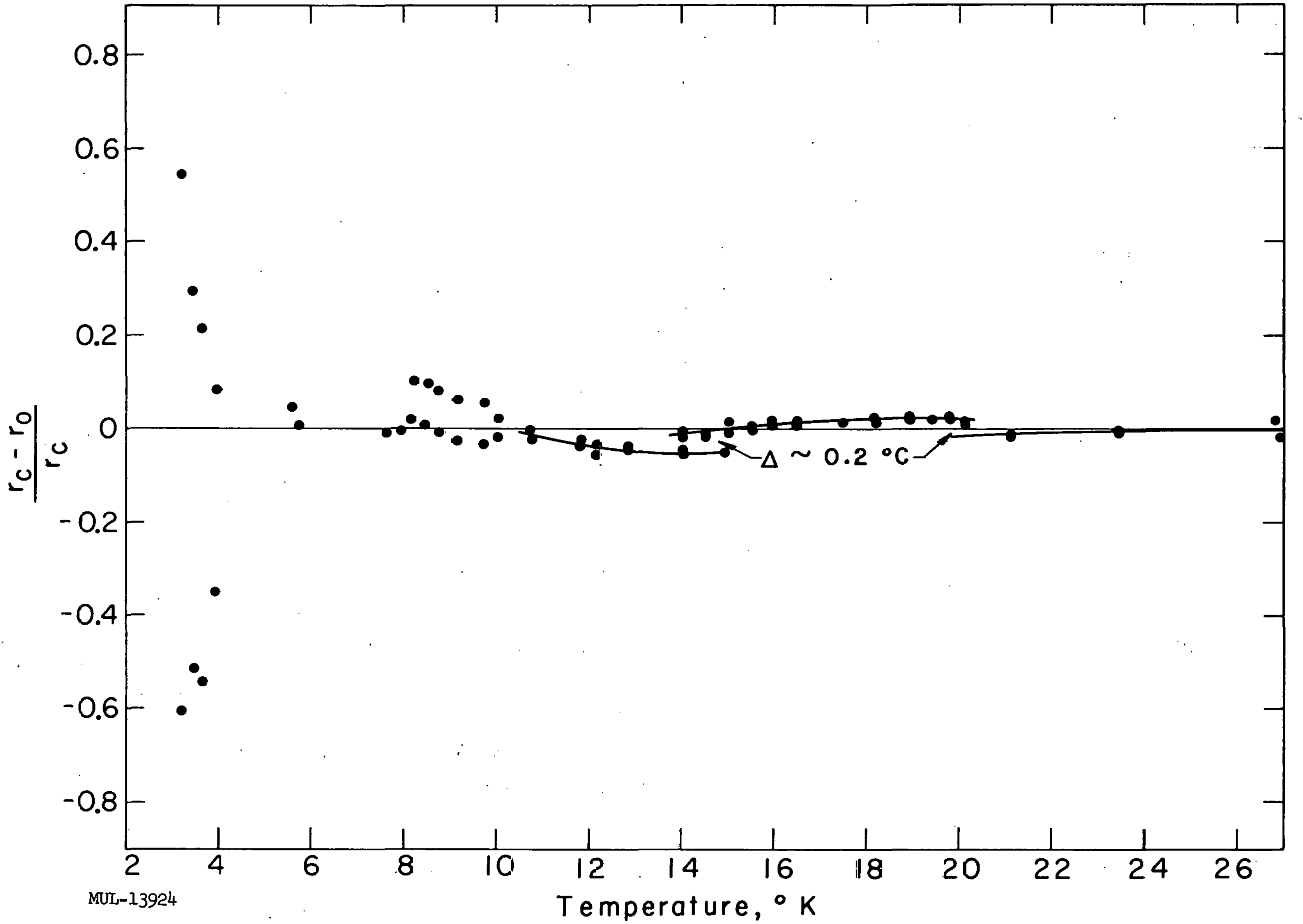

Figure 5. Fit of equation (2) on raw data for aluminum wire sample No. 8, $R_{300}/R_{4.2}$ = 1834

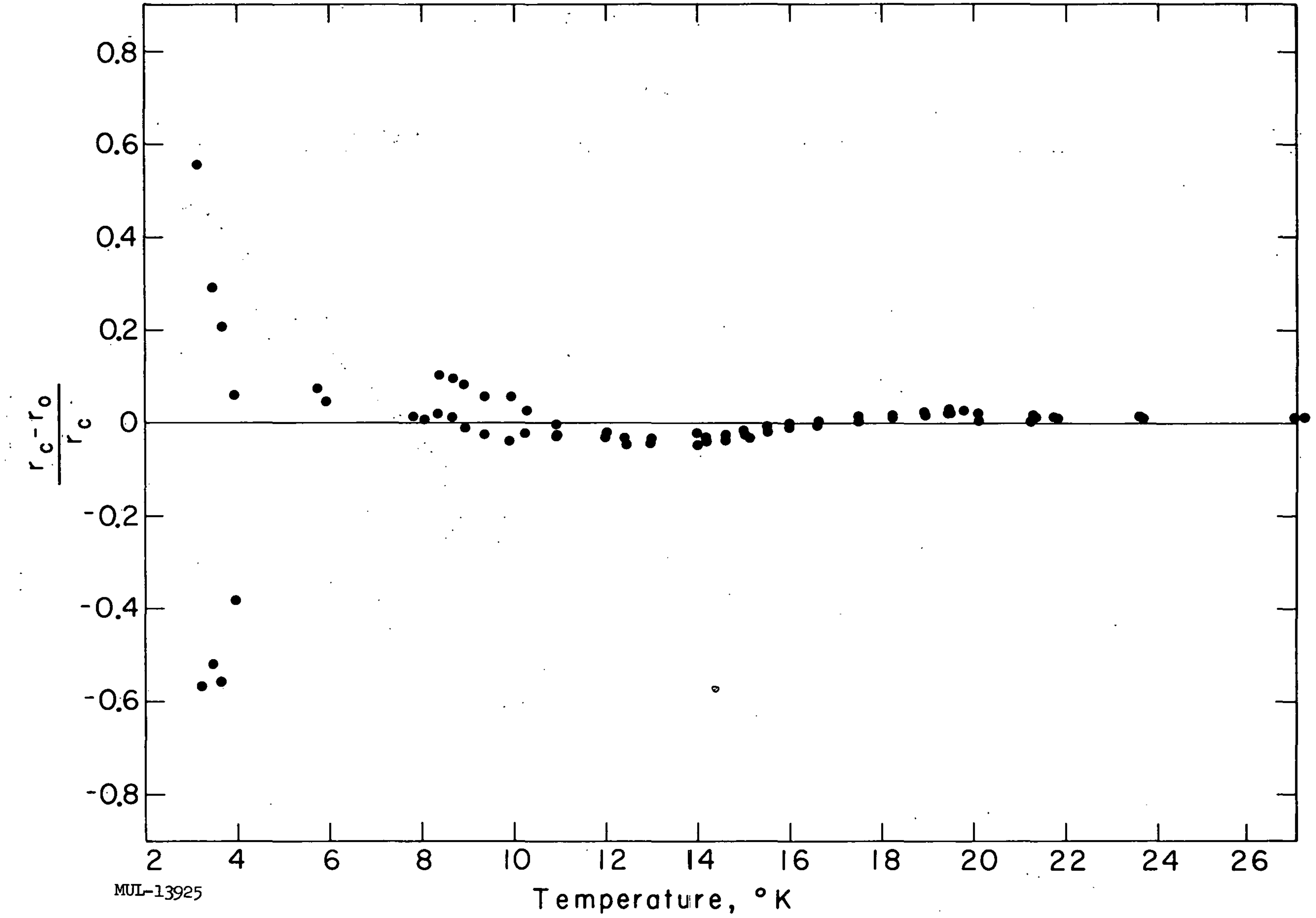

Figure 6. Fit of equation (2) using corrected temperatures, aluminum wire sample No. 8, $R_{300}/R_{4.2}$ = 1834

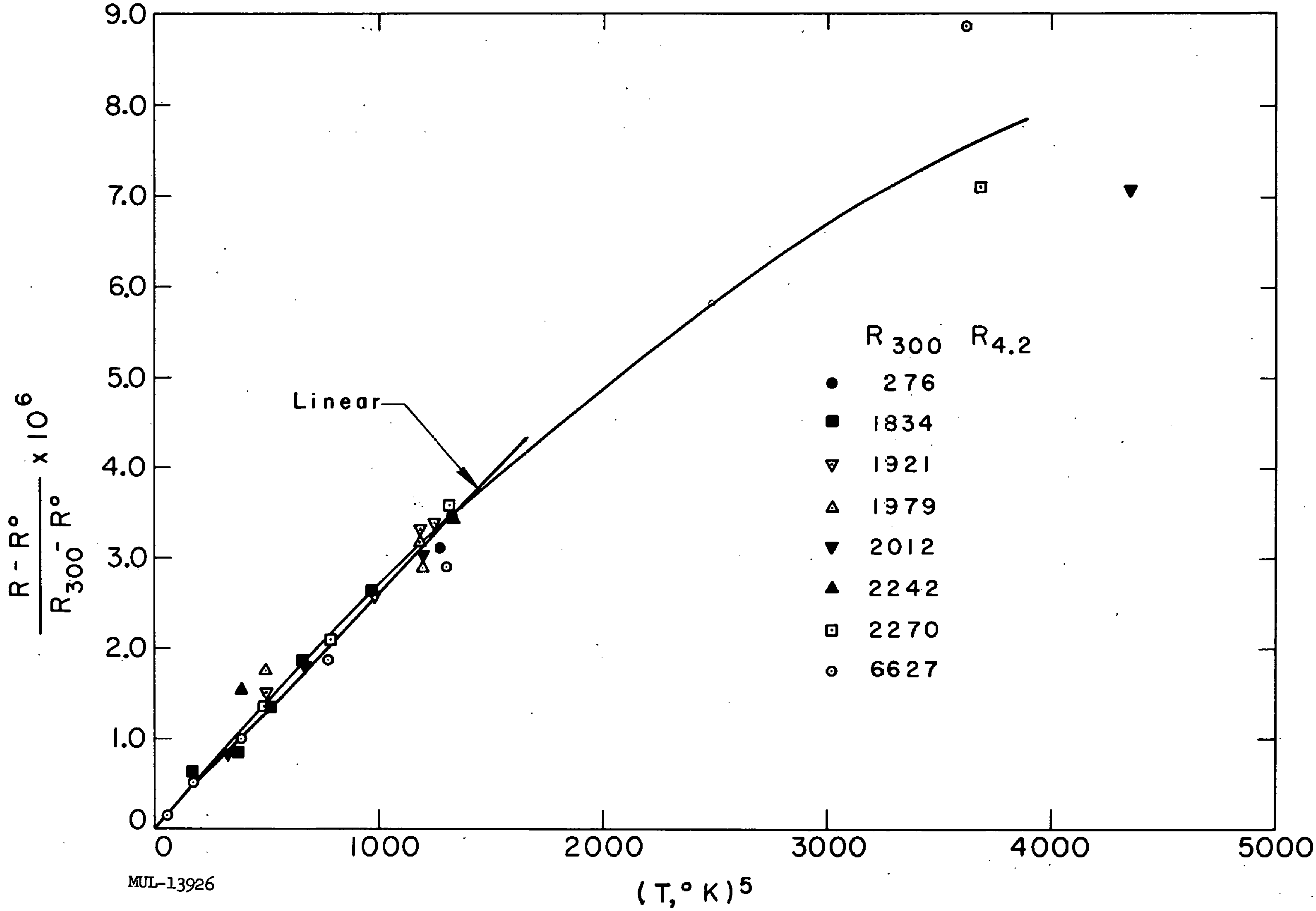

Figure 7. Reduced resistance vs T^5

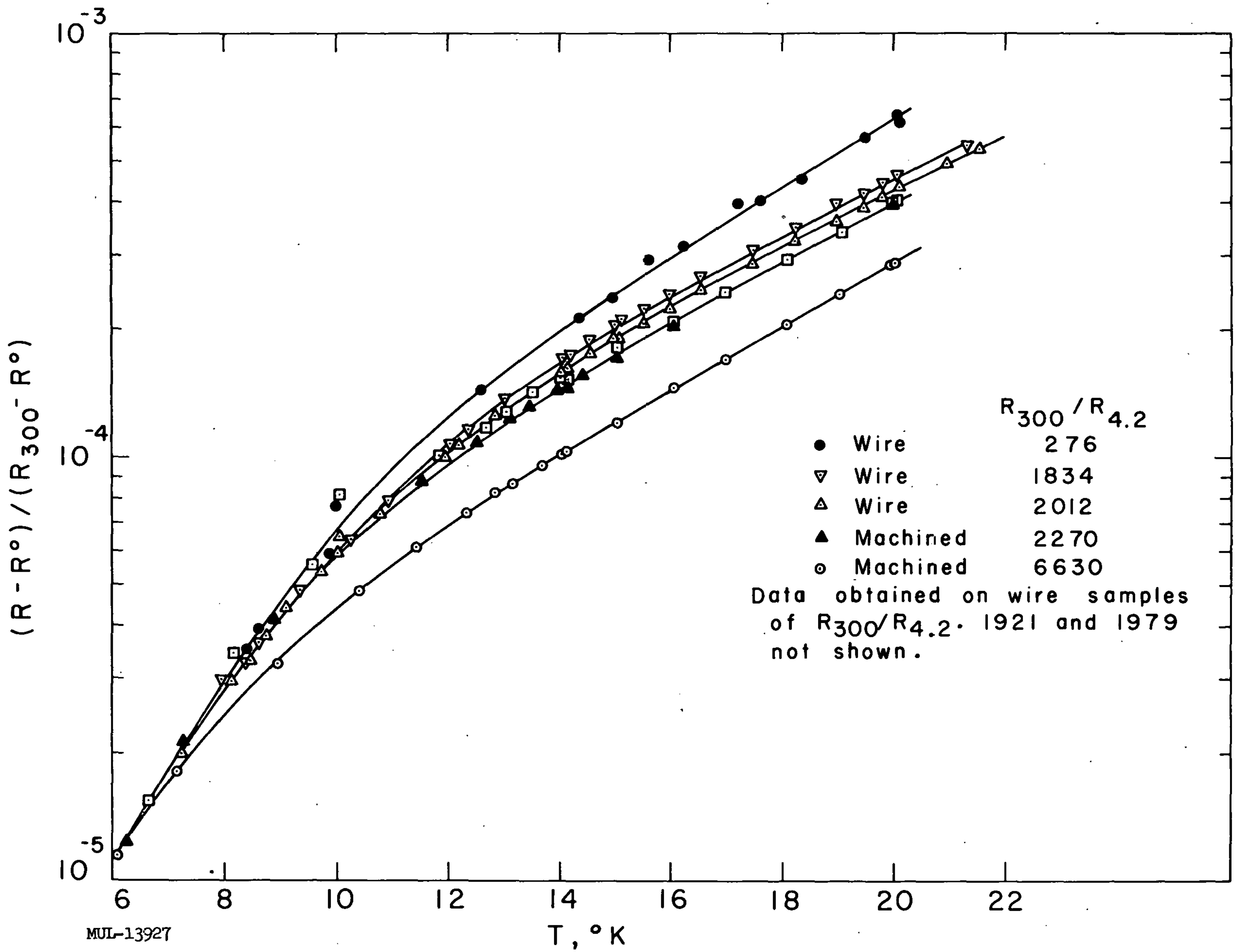

Figure 8. Reduced resistance vs temperature

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