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Effect of Recovery on the Recrystallized Grain-Size of High Purity Aluminum

Rodney L. Helterline

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A Thesis

Submitted to the Department of Metallurgy

in Partial Fulfillment of the

Requirements for the Degree of

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AN ANALYSIS OF ANNEALING

When a cold-worked metal is annealed, its physical properties change as a result of a new grain structure. The annealing treatment is divided into three stages according to the changes that occur in the distorted metal: (1) recovery, (2) recrystallization, and (3) grain growth.

Recovery occurs without any observable change in microstructure but it is accompanied by a decrease in residual stress and electrical resistance. During the recovery process, the other physical properties, other than electrical resistance, remain substanially unchanged.[12]

As the annealing temperature is raised, a point is reached at which cold-worked metals begin to soften and to regain their plasticity. The microstructure also changes markedly during this stage of annealing. The distorted, elongated grains produced by the previous cold working are gradually replaced by a number of small grains that continue to grow at the expense of the strained grains until the latter disappear and the structure consists entirely of the newly formed grains. This phenomenon is called recrystallization. It is a combination of both nucleation and grain growth reactions. For most purposes, this is the important stage of annealing. The new grains are essentially strain-free and their average size depends on a number of factors including the amount of cold working done, the annealing temperature used, the rate of heating, the amount of impurities, and other contributing factors.

The grain size at the completion of recrystallization depends principally on the degree of deformation prior to the annealing.[12] The higher the degree of cold work, the lower will be the temperature necessary to produce complete recrystallization in a given annealing period, and the smaller will be the grain size.

The temperature at which a cold-worked metal begins to recrystallize is lowered by increasing the time of annealing. An increase in the amount of cold work and the presence of impurities also lower the recrystallization temperature.

The recrystallization of a cold-worked metal is said to be complete when the distorted grains are entirely replaced by the newly formed grains. However, metals continue to become softer and more ductile as the annealing temperature is raised beyond that necessary to complete the recrystallization. This is due to grain growth, which only occurs after recrystallization is completed, and provided that the recrystallized grain size is reasonably small.

When grain growth occurs above the recrystallization temperature, the grain size attained depends principally upon the temperature and secondarily upon the time of annealing. At any given annealing temperature above recrystallization, the grains grow to a size that is characteristic of the temperature, and no perceptible growth occurs thereafter. This characteristic size increases rapidly with increasing temperature.

OBJECT OF INVESTIGATION

A number of investigators have observed that prior recovery does have an effect on recrystallization, but no systematic and thorough investigation has been made or at least no published results are available in the literature. Indications are that recovery in commercial aluminum retards tha rate of subsequent recrystallization.[1] The decrease in recrystallization rate has been attributed to a decrease in the rate of nucleation, with no change in the rate of growth. The purpose of this investigation was to determine quantitatively the effects of time and temperature of the recovery period on the final recrystallized grain size of high purity aluminum.

MATERIAL SELECTED FOR INVESTIGATION

High purity aluminum, supplied by the Aluminum Company of America was used in this investigation. The metal had the following composition:

Silicon	0.002%
Iron	0.001%
Copper	0.003%
Magnesium	0.001%

The selection of high purity aluminum was due to the availability of accurate recrystallization values. Anderson and Mehl's work established the time and temperature conditions for recrystallization of high purity aluminum for elongations of 5, 10, and 15%. They also determined the "activation energies" of recrystallization for the same elongations. All of these values were later checked and confirmed by Griswold[7].

SELECTION OF RECOVERY TIMES AND TEMPERATURES

The recovery times and temperatures were selected so that their combinations would not produce recrystallization. Assuming recovery times of 2.4, 24, and 240 hours, the corresponding temperatures were calculated by using Anderson and Mehl's data[1] in the equation on page 12. These temperatures for 240 hours are 313°C, 295°C, and 278°C for 5, 10, and 15% elongations, respectively. They represent the lowest temperatures when combined with the corresponding times that will produce recrystallization. Any lower temperatures will be in the recovery zone, and, therefore, 150°C, 75°C, 8°C were chosen for the recovery temperatures.

To further justify the chosen recovery temperatures, Hultgren's[10] recrystallization data for three purities of severely deformed aluminum was plotted. These recrystallization temperatures for 24 hours are as follows:

99.9986% Al	50°C
99.996% Al	185°C
99.98% Al	225°C

From the curve which was plotted from these values, 99.993% Al, which was used in this investigation, was shown to recrystallize at 200°C. This temperature was reduced to 184°C when the time was lengthened to 240 hours. Thus, the chosen recovery temperatures were in the recovery zone and would not produce recrystallization.

METHOD OF INVESTIGATION

To determine the effect of recovery on the recrystallized grain size, the author subjected a number of aluminum specimens to a recovery treatment, and then recrystallized them without any unnecessary grain growth. Without any prior recovery treatment, all of the recrystallized specimens should have the same grain size, therefore, any variation in the grain size can be attributed to recovery.

Tensile Bars

The high purity aluminum, which was received as a 3/4 in. rod, was cold swaged down to a $\frac{1}{2}$ in. rod from which tensile bars were machined. These tensile bars were 3 in. in length with a $\frac{1}{4}$ in. x 1 $\frac{1}{2}$ in. gage section (Figure 1). The gage sections of all bars were polished, first with 1/0 and then with 3/0 emery cloths, to insure both uniform diameters and smooth surfaces. The sections were accurately measured with a micrometer and polished down to a variation of less than ± 0.001 inches.

Initial Anneal

Following polishing, the tensile bars were placed in a furnace at 800°F for 15 minutes to provide a uniform annealed grain structure. A sample of the aluminum in the as-swaged condition was also given this heat treatment, from which the original grain size was obtained.

Deformation

Uniform deformation was obtained by elongating the tensile bars in a tensile apparatus used by Griswold.[7] This apparatus consisted

FIGURE 1. TENSILE BAR

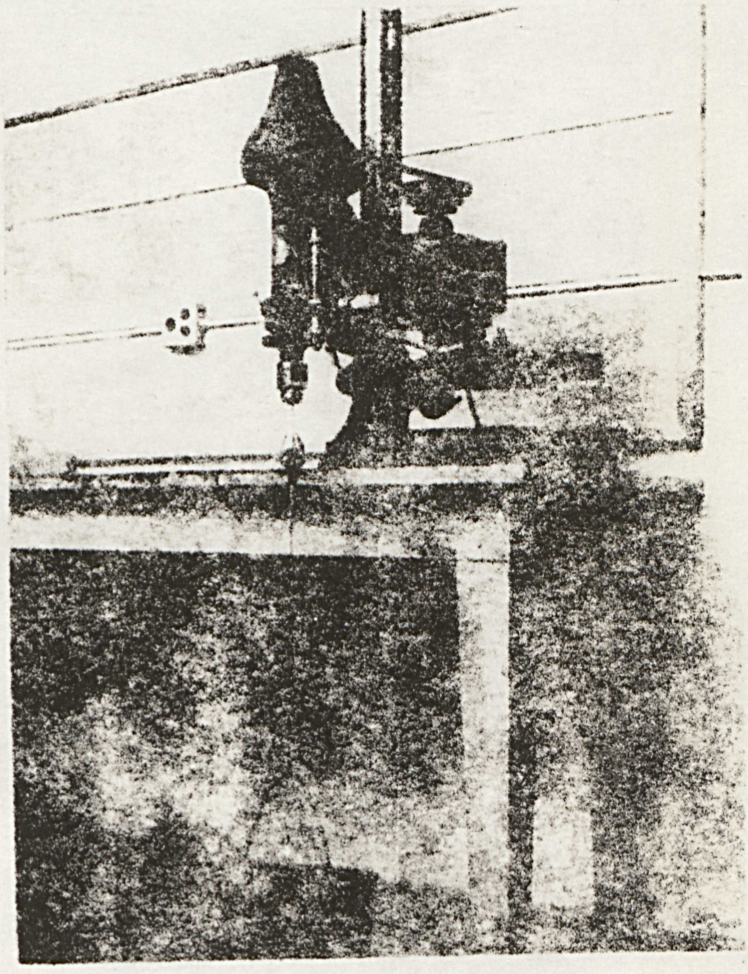


FIGURE 2. TENSILE APPARATUS

of a drill press chuck, which acted as the upper or fixed specimen holder, and a lathe chuck, to which a load could be attached for a movable specimen holder (Figure 2). The upper end of the tensile bar was fastened into the chuck of the drill press, which was clamped to a bench and rotated 90° from a working position about the vertical support column. This allowed the driven part of the drill press to hang free over the bench. The lower end of the tensile bar was fastened in the lathe chuck, which, in turn, was connected to a bucket by a piece of piano wire. Elongation was effected by slowly pouring test lead into the bucket.

The amount of deformation produced was determined with the aid of a pair of dividers. Two very fine points, 1.25 in. apart, were inscribed on the surfaces of the gage sections of the tensile bars. The elongation was carried out until the distance reached a value which corresponded to the desired elongation. The necessary extension to provide a certain elongation percentage was calculated by the use of the following equations. The diameters were obtained by micrometer measurement and from these a percent reduction in area might easily be calculated. The results are shown in Table I.

$$L_f - L_i = \Delta L$$

$$\% \text{ elongation} = \frac{\Delta L}{L_i} \times 100$$

where L_i = Initial Length

L_f = Final Length

ΔL = Change in Length

Table I

Elong.	Initial Dia.	Final Dia.	Initial Dist.	Final Dist.	Wt. to Pull
5%	0.225	0.222	1.25	1.31	208#
10%	0.230	0.220	1.25	1.31	220#
15%	0.209	0.197	1.25	1.31	232#

Tensile Bar Cutting

Immeadiately after elongation, the gage portion of each tensile bar was cut into 9 sections. To minimize deformation of the aluminum during the cutting operation, the author employed two precautions. The first, which Griswold also used,[7] was to employ two blocks of balsa wood, from each of which, a semicircular trough was cut. The tensile bar gage section could be held firmly in the troughs when the two blocks of wood were clamped together. Secondly, a Jewler's saw, which had a very fine, sharp-toothed blade, was then used to cut the specimens from the gage sections. Also, to prevent heating, a steady stream of water was applied to the specimens while they were being sawed.

Recovery Treatment

As soon as the tensile bars were cut, the specimens were put in furnaces (Figure 3) which had been heated previously to the recovery temperature range (75°C, 150°C), or in the freezing tray of a refrigerator (8°C). The various temperatures and times used are given in Table II. They were selected so as to give a wide range of time and temperature conditions. Since each tensile bar was cut into 9 sections,

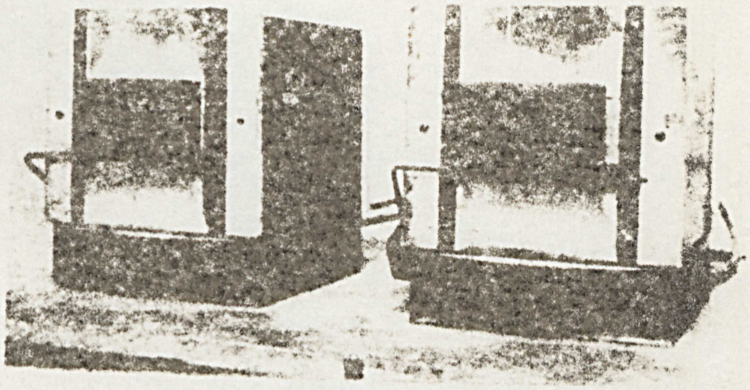


FIGURE 3. . RECOVERY FURNACES AT 75° and 150°C

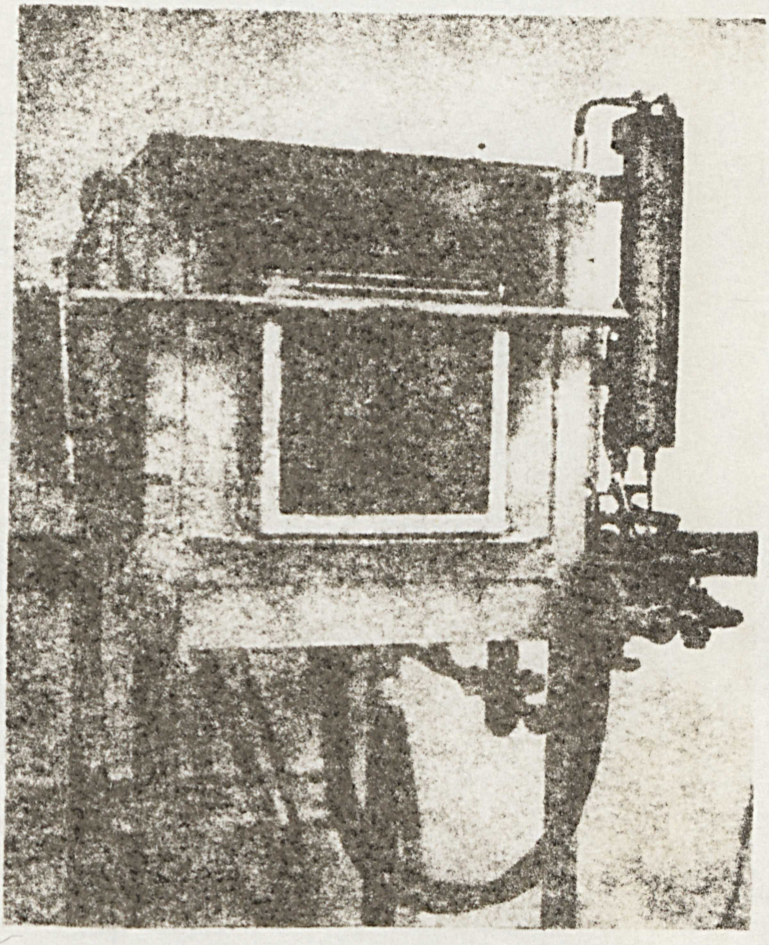


FIGURE 4. . RECRYSTALLIZATION FURNACE

3 sections could be treated at each temperature, and one could be removed at each different time indicated in Table II.

Table II

Elongation	Temperature (°C)	Time (hrs.) t_1	t_2	t_3
5%	8	2.4	24	240
	75	"	"	"
	150	"	"	"
10%	8	"	"	"
	75	"	"	"
	150	"	"	"
15%	8	"	"	"
	75	"	"	"
	150	"	"	"

Recrystallization

Following the recovery treatments, the specimens were put directly into another furnace for recrystallization (Figure 4). The combination of temperature and time for complete recrystallization without unnecessary grain growth is a function of the amount of elongation. The length of time required for complete recrystallization has been calculated by Anderson and Mehl[1] for a temperature of 350°C. However, at this temperature, a short heating time was required for those experiments in which the elongation was 10 and 15%. It was thought that lenghtening the time to approximately 4 hours would reduce error due to the specimen coming up to temperature. This was effected by decreasing the temperature of recrystallization. On the other hand, the long time required for recrystallizing the 5% elongated specimens was reduced to approximately 4 hours by raising the recrystallization temperature.

These new values are calculated by explicitly integrating a logarithmic form of the Arrhenius equation ($R = Ae - \frac{Q}{RT}$) with respect to time:

$$\log \frac{t_2}{t_1} = \frac{Q}{2.303R} \frac{(T_2 - T_1)}{(T_2 T_1)}$$

where Q = "activation energy" for recrystallization

R = molar gas constant

t_1 = time required to effect complete recrystallization at T_1

t_2 = " " " " " " " T_2

T_1 = temperature for time t_1

T_2 = " " " t_2

The value Q and the recrystallization times at 350°C as determined by Anderson and Mehl[1] are listed in Table III. An example of the calculation to determine the new recrystallization temperature is shown below. The recrystallization times and temperatures are listed in Table IV.

Calculation for 5% elongation

t_2 = 500 min. = annealing time at 350°C

t_1 = 219 min. = desired time of annealing

T_2 = 350°C or 623°K

Q = -64,300 cal.

$$\log \frac{500}{219} = \frac{-64,300}{2.3 \times 1.987} \frac{(623 - T_1)}{(623\ T_1}$$

$$0.357 = -14,060 \frac{(623 - T_1)}{(623\ T_1)}$$

$$13,839\ T_1 = 8,760,000$$

$$T_1 = 633°K \text{ or } 360°C$$

Table III

Elongation	Q (cal)	Annealing time at 350°C
5%	-64,300	500 min
10%	-58,800	135 min
15%	-53,050	36.5 min

Table IV

Elongation	Temperature	Time
5%	360°C	219 min
10%	343°C	232 min
15%	327°C	189 min

Polishing and Etching

When recrystallization was completed, one flat surface of each specimen was ground on 1/0, 2/0, and 3/0 emery papers. A saturated solution of paraffin in kerosene was used on the emery paper to decrease the metal pickup on the emery paper.

A galvanic etch, as used by Servi[12], was first tried. However, an acid etch, developed by Barrett and Levenson[2], proved to be more satisfactory. It consisted of a mixture of 9 parts HCl, 3 parts HNO_3, 2 parts HF, and 5 parts H_2). The etching time varied from 25 to 35 seconds.

Photographing Procedure

The etched samples were photographed at 12.5x so that an image

of the entire specimen surface could be included on one 3 1/4 in. by 4 1/4 in. film so that all the grains could be measured, if desired. Since the low magnification could not be obtained with a standard metallograph, a 72mm B and L Micro Tessar lens was attached to the bellows of a B and L Metallograph from which the shutter and adapter had been removed. Illumination of the specimens was done by means of two spotlights, one on each side of the specimen to be photographed. It was found that better grain definition could be obtained with illumination from one side only; therefore, two exposures were taken of each specimen -- one with each light. A 20 second exposure with a subsequent 2 minute developing time in Kodak DK 50 gave satisfactory macrophotographs with Kodak Contrast Process Ortho film.

Grain Area Measurements

It was decided that the most accurate method to determine the average grain area was to measure the areas of a series of individual grains. To increase the accuracy of the measurements, each negative was placed in an enlarger, and the grains were measured on the projected image. The negatives were enlarged 4.14x, and hence the projected grains were at 51.8x. Area measurements were made by means of a planimeter.

To obtain a representative sample, all the grains in a quadrant of the circular area projected were measured. Grain size measurements were made on each of the two exposures made of each specimen. The average grain size from each negative was found, and then both these

values were averaged to give an average final grain size. This value, in square inches, was divided by 51.8 and mutiplied by 6.45 to give the actual grain size in square centimeters. These final grain sizes are shown in Table V.

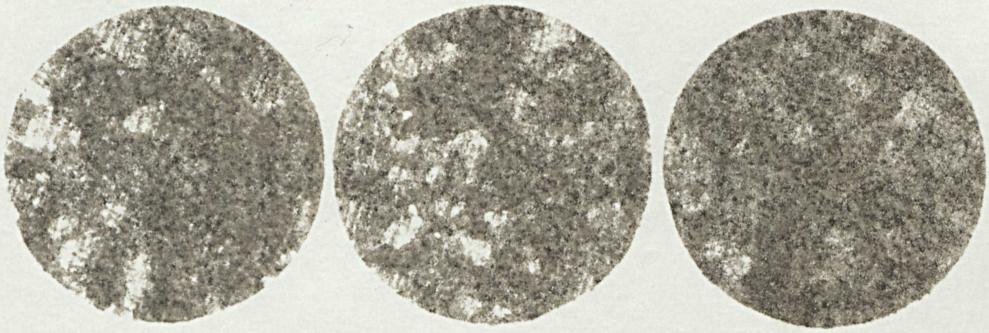
RESULTS

The results are clearly shown in Table V and by the photographs on the following pages. Three graphs, in which the grain size is plotted against the log of the recovery time, are included to show the effect of recovery time on the final recrystallized grain size.

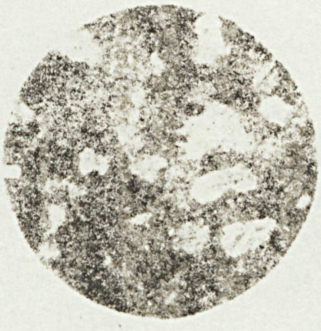
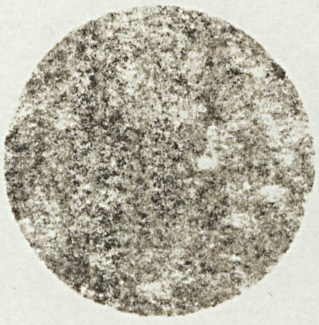
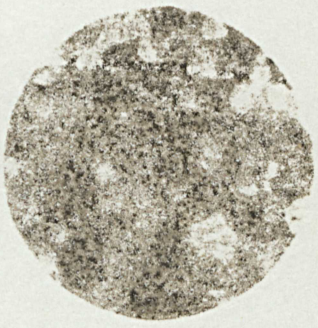
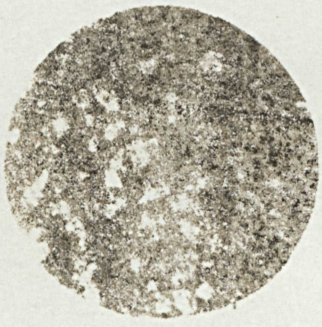
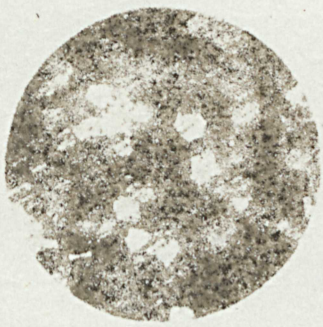
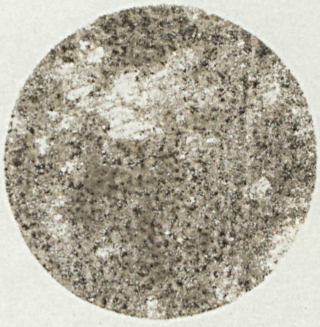
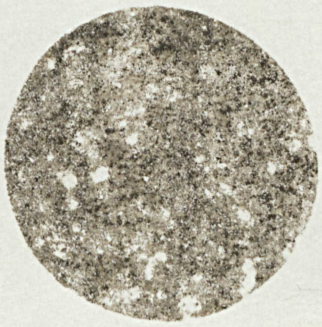
Table V

Deformation	Recovery Temp.	Time (hr)	Grain Size (sq.cm.)	No. of Grains per sq. cm.
5%	150°C	240	.0236	42.4
		24	.0294	34.0
		2.4	.0364	27.5
	75°C	240	.0266	37.6
		24	.0193	51.8
		2.4	.0652	15.3
	8°C	240	.0588	17.0
		24	.0209	47.8
		2.4	.0181	55.2
10%	150°C	240	.0496	20.2
		24	.0305	32.8
		2.4	.0127	78.8
	75°C	240	.0300	33.3
		24	.0326	30.7
		2.4	.0168	59.5
	8°C	240	.0331	30.2
		24	.0333	30.0
		2.4	.0128	77.6
15%	150°C	240	.0205	48.8
		24	.0187	53.5
		2.4	.0173	57.8
	75°C	240	.0382	26.2
		24	.0214	46.8
		2.4	.0185	54.1
	8°C	240	.0223	45.9
		24	.0312	32.1
		2.4	.0208	48.1
Initial Grain size after annealing at 800°F for 15 min.			.0307	32.5

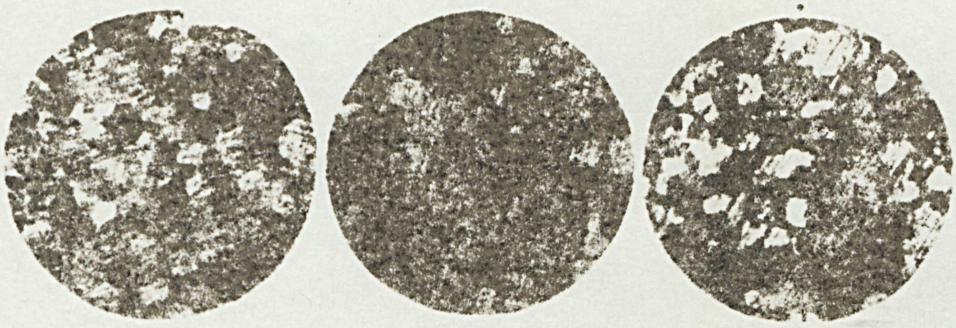
ELONGATION 5 o/o RECRYSTALLIZATION TREATMENT----219 MIN. AT 360°C

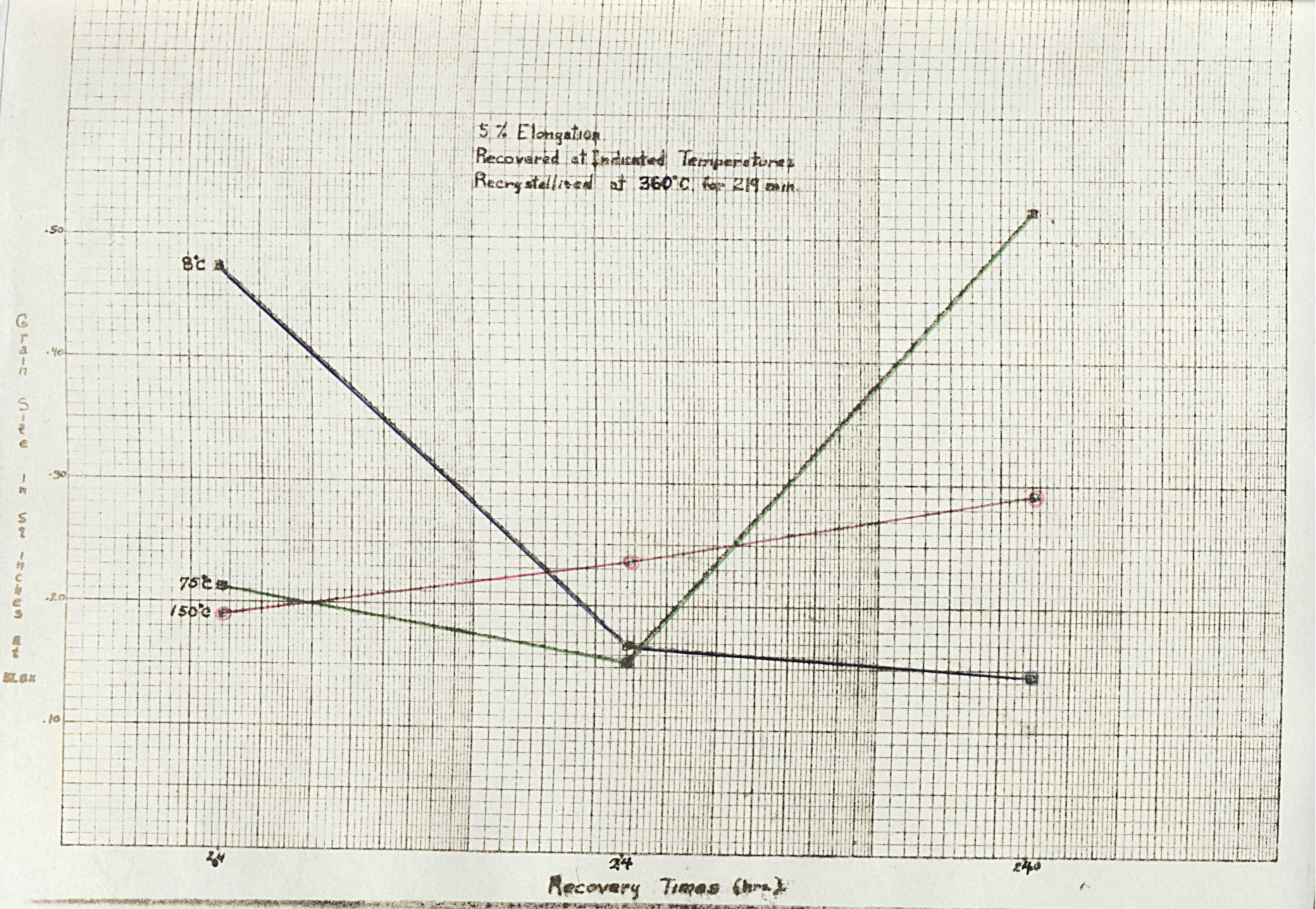
RECOVERY TEMP. \ RECOVERY TIME	2.4 HRS.	24 HRS.	240 HRS.
150°C			
75°C			
8°c			

ELONGATION 10 o/o RECRYSTALLIZATION TREATMENT-----232 MIN. AT 343°C

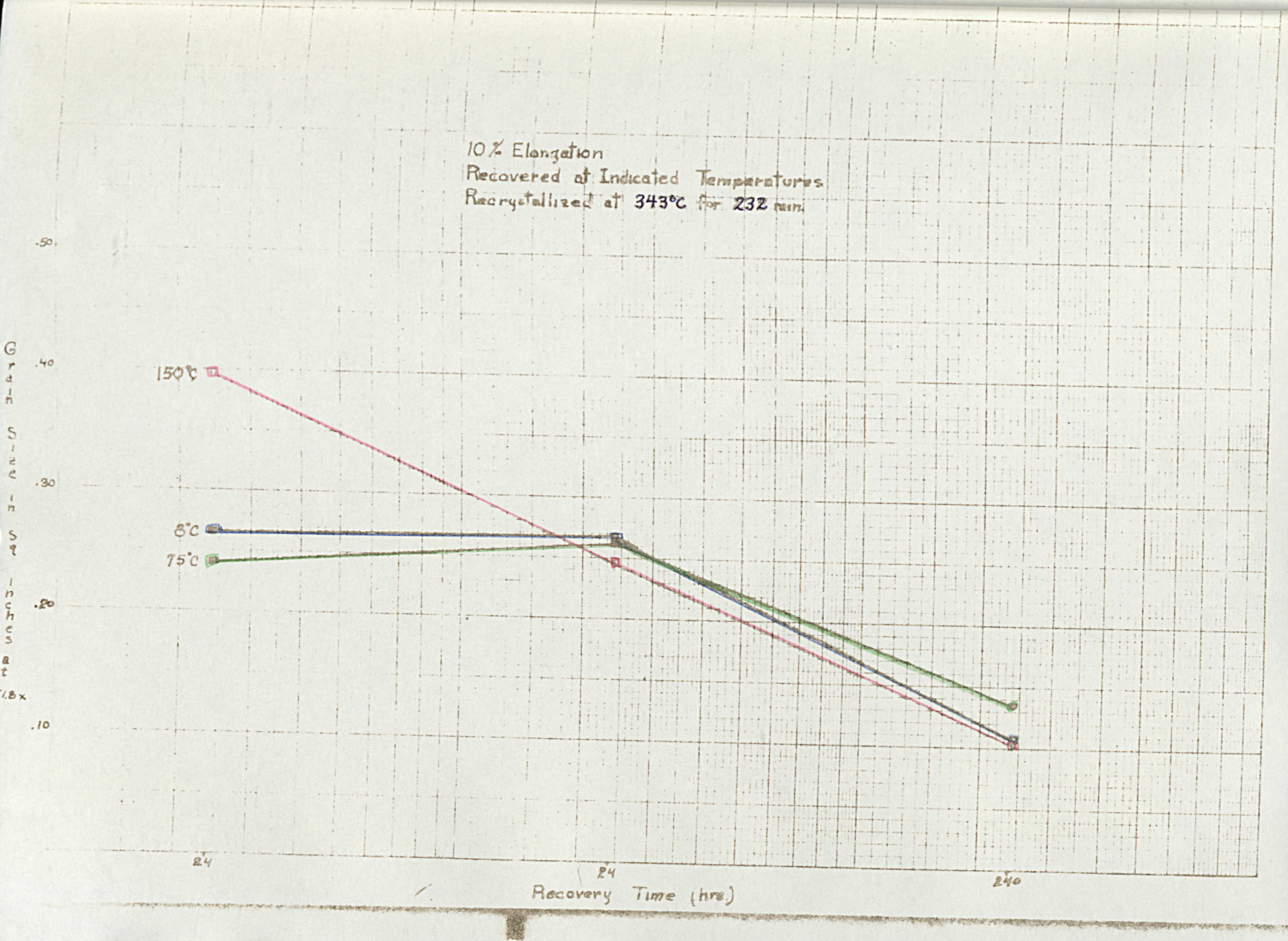
RECOVERY TIME	2.4 HRS.	24 HRS.	240 HRS.
150°C			
75°C			
8°C			

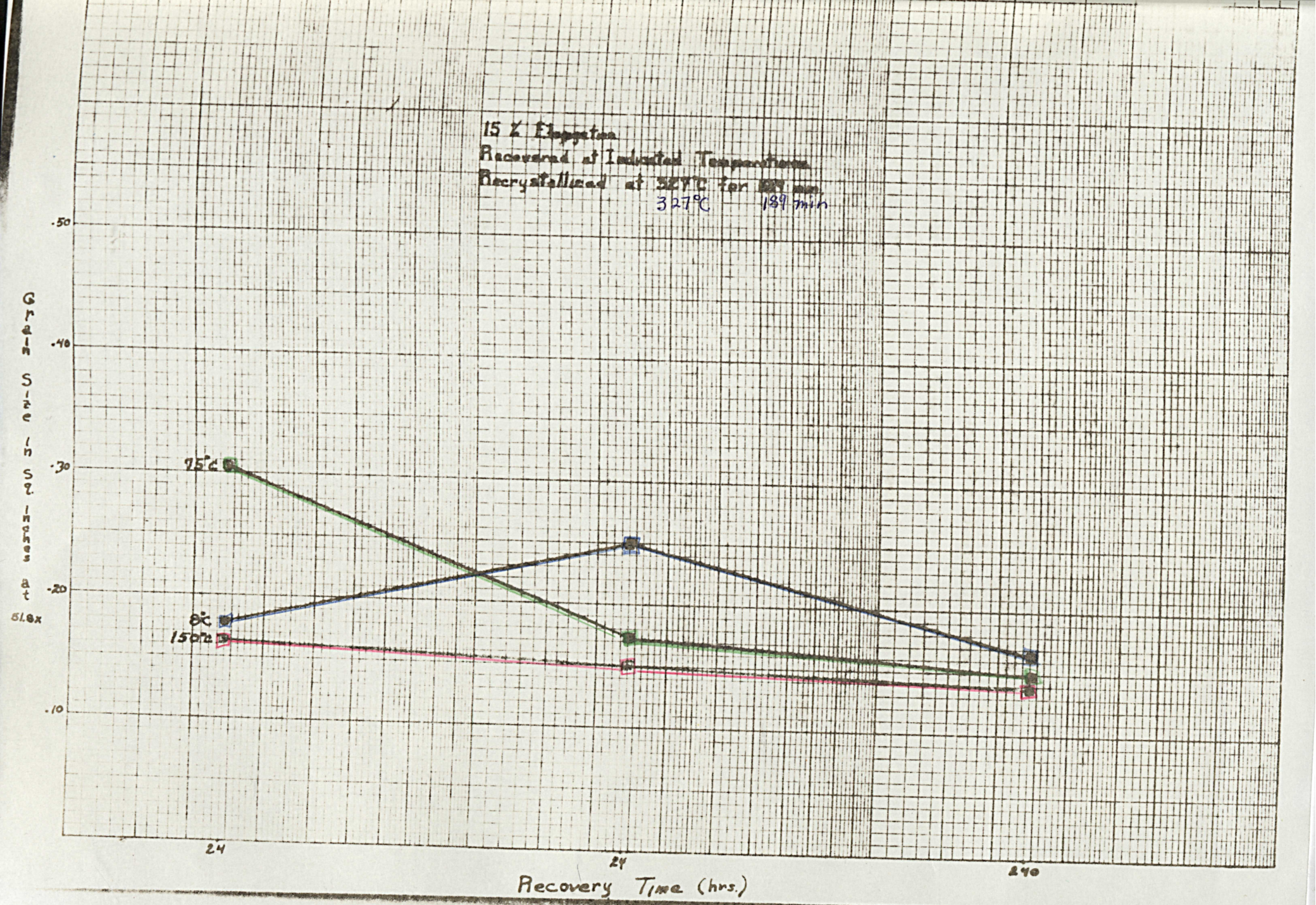
ELONGATION 15 o/o RECRYSTALLIZATION TREATMENT---189 MIN. AT 327°C

RECOVERY TEMP. \ RECOVERY TIME	2.4 HRS.	24 HRS.	240 HRS.
150°C			
75°C			
8°C			


5 % Elongation
Recovered at Indicated Temperatures
Recrystallized at 360°C. for 219 min.
8°C
75°C
150°C
.50
.40
.30
.20
.10
Grain Size in Sq. Inches at 87.5X
24
24
240
Recovery Times (hrs.)

10% Elongation
Recovered at Indicated Temperatures
Recrystallized at 343°C for 232 min.




15 % Elongation
Recovered at Indicated Temperatures
Recrystallized at 327°C for 139 min
327°C 139 min
.50
.40
.30
.20
.10
Grain Size in Sq. Inches at 51.8x
75°C
8°C
150°C
24
24
240
Recovery Time (hrs.)

DISCUSSION

The plots of recrystallized grain size vs. log recovery time are somewhat difficult to analyze because the number of experimental points is so limited. A well-defined trend in evident only in those specimens recovered at 150°C. In these specimens, the progression seems to be toward a finer recrystallized grain size with time for the samples deformed 10% and 15% by elongation and toward a coarser recrystallized grain size with time in the samples deformed 5%. In all of these specimens, a straight line can be used to best represent the locus of the experimental points.

For the specimens elongated to 10 % and 15% and recovered at 8°C and 75°C, the trend in the samples is also generally toward a finer recrystallized grain size as the time of recovery is increased. For the specimens elongated to only 5%, the grain size decreases with time only in the case of the samples recovered at 8°C. The samples recovered at 75°C show a slight decrease in grain size after 24 hours and a large increase after 240 hours recovery and subsequent recrystallization.

Contributing factors which make it necessary to consider the results only in terms of the above generalizations are the following:

1. As mentioned before, a small amount of impurity will influence the recrystallization conditions very markedly, and local small variations in composition might well be suspected.

2. On deformation of the specimens there was a tendency for the weight bucket to twist slightly with the addition of the shot. This

would cause a strain in excess of that indicated by the elongation and may also result in non-uniform straining and consequent grain size variation.

3. The grain size was carefully measured and the values obtained are considered dependable from the aspect of measurement. However, it must be recognized that the macro-etching technique used may have resulted in a loss of grain boundary definition and possibly a consideration of two grains as a single one in some instances. This is thought to have been averaged out for most specimens and is probably a relatively minor factor.

RECOMMENDATIONS FOR FURTHER INVESTIGATION

As can be seen from the results of this investigation, much work needs to be done on the subject of annealing and particularly that of recovery. Experiments of this nature are very worthwhile because of their practical importance. If further investigations are to be conducted, the author would like to offer the following suggestions.

1. The elongation apparatus should be constructed so as to prevent the twisting of the tensile bars as a result of the weight bucket revolving back and forth.

2. An etch should be used that will give good grain definition. A completely satisfactory and reliable etch was not found until after the conclusion of this experiment. It was an electrolytic etch used by Hone and Pearson[9] and Sperry[13], and seemed to offer a very good technique for aluminum electropolishing and electroetching.

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