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Preparation of copper single crystals by means of powder metallurgy methods

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PREPARATION OF COPPER SINGLE CRYSTALS BY MEANS OF POWDER METALLURGY METHODS

BY

WEI-KUNG CHANG

A

THESIS

submitted to the faculty of the

SCHOOL OF MINES AND METALLURGY OF THE UNIVERSITY OF MISSOURI

in partial fulfillment of the work required for the

Degree of

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Approved by Daniel S Eppelsheimer
Professor of Metallurgical Engineering

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I

Introduction

Since different mechanical properties of metals result from different grain size became known, the metallurgists have been interested in finding out the true nature of the grain boundaries. Consequently, several different theories concerning the mechanism of recrystallization, grain growth, and the structure of metals were advanced and studied. These threw much light on the difference of properties between polycrystalline metals and single crystals of the same metals. The single crystals of metals were studied because they enabled the investigator to determine the important points of metallic behavior. The first work on single crystals was rather obscure and limited to isolated, though significant, observations on native metals and on large grains which were removed from severely coarsened metal parts.

The aim of the present research was to investigate the possibility of preparing the single crystals from copper powder by employing the powder metallurgy technique. However, the principal differences between cast and sintered metals are the relative weakness of the adhesive forces between particles and the presence of pores between them in the case of powder compacts. The factors controlling grain growth and grain size for powder compacts depend not only upon the amount of deformation, temperature and time of annealing, but also depend upon the amount of compressing pressure and the sintering conditions of powder compacts.

This research consisted of three separate experiments delineated briefly as follows:

(1) Compressed copper powder flats ------ A series of compressed copper powder flats were prepared with varying amounts of compressing pressure, and sintered under different time and temperature. These samples were then followed cold worked in compression and annealed. This working and annealing was repeated eight times. Specimens with a final grain size almost equal to the thickness of the test piece resulted from the use of compressive loads varying between 8 to 12 tons per square inch. In addition, the volume change and density change were measured.

(2) Copper wire ------ For the purpose of comparing the difference of cast metal and powder compacts and trying to determine the faverable temperature for grain growth of copper, pure copper electric wire with 1/16 inch in diameter was used. The procedure consisted of first, annealing the copper wire; second, elongating in tension to produce a known cold deformation; and third, annealing for recrystallization and grain growth. It was found that 2% elongation gave the largest crystals about 1/16 inch in thickness when annealed at 950°C (1742°F) for five hours.

(3) Compressed copper powder tensile bars ------ The compressed copper powder tensile bars, which were prepared by employing the optimum compressing pressure range determined from the work on the copper flats, were sintered at 950°C for five hours, deformed by 2% elongation, and annealed at 950°C for thirty hours. The resulted sample was not a single crystal, though it exhibited a very large grain size.

II

Review of the Literature

A. Principles of recrystallization and grain growth

1. Recrystallization ——— Recrystallization[1] means that on heating a cold worked metal to a sufficiently high temperature or for a sufficiently long time at a somewhat lower temperature new grains appear and grow until ultimately a new grain structure has completely replaced the old.

Metals which have been strained by cold working are not stable, but tend to revert to a strain-free state. From the standpoint of energy, in the deformation of a metal, we have seen that slip takes place along definite crystallographic planes in definite directions and that slip results in a distortion of the slip planes and grain boundaries, with an accompanying increase in internal energy. We should expect that energy increase to be located on the slip planes and in the grain boundaries, as these are the points at which the actual deformation takes place. Thus at definite points throughout the deformed metal there are localized regions of high-energy content. The amount of deformation determines the number and magnitude of these high-energy areas. If the temperature of the metal is raised the energy content of the entire piece is raised. When the temperature is reached at which the localized areas have sufficient energy to overcome the rigidity of the distorted lattice, they give up their energy as a heat of recrystallization, and form nuclei

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(1) Mehl, Robert F., Recrystallization. ASM, Metals Handbook, p. 261.

for the growth of new, stress-free crystals from the surrounding distorted metal. The number of nuclei precipitated depends upon the amount of previous deformation. The number of nuclei also depends upon the temperature of annealing; the higher the temperature, the more areas will be raised to an energy level sufficiently high to precipitate nuclei. The first point to precipitate a nuclei for the formation of a single crystal should be the area in which the concentration of energy due to cold work is the highest. Under favorable conditions, the nuclei should grow to form a single crystal from a large volume of polycrystalline material.

The factors controlling the final grain size after recrystallization can be summarized as follows:

1. The amount of cold deformation,
2. The temperature to which the cold worked metal is heated,
3. The time for which the metal is held at this temperature, and
4. The original grain size before cold working.

Mehl(2) states that the following fundamental laws of recrystallization can be recognized: (1) Recrystallization occurs at a higher temperature the smaller the degree of deformation or, more accurately, the smaller the degree of strain hardening. (2) Increasing time of annealing displaces recrystallization to a lower temperature. (3) The final grain size is chiefly dependent upon the degree of deformation and to lesser degree on the annealing temperature, the final grain size is the larger, the smaller

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(2) Mehl, Robert F., Recrystallization, ASM Metals Handbook, 1948, p. 261

the degree of deformation. (4) The larger the original grain size the greater the amount of cold deformation to give equivalent recrystallization temperatures and times. In addition to these major factors, it should be noted that (a) the rate of heating to the recrystallization temperature is a further variable, and (b) the amounts of cold work to give equivalent strain hardening vary with the temperature at which deformation is performed.

2. Grain growth

By the definition in the ASM Metals Handbook, grain growth means an increase in the grain size of the metal. According to Beck(3), grain growth can be classified as follows:

1. "Normal grain growth ---- The type of grain growth occurring in uninhibited Cu-Be alloys has been most frequently and best investigated in brass. This type has been designated as normal grain growth."
2. "Abnormal grain growth ---- Grain growth occasionally to the phenomenon of critical recrystallization which also results in large grains by different mechanism."
3. "Continuous grain growth ---- Grain growth in uninhibited metals and alloys is the continuous increase of the average diameter with increasing time and temperature of annealing, and the grain size is relatively uniform at all temperatures."

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(3) Beck, Paul A., Grain growth in high purity Al and Al-Mg alloy, A.I.M.E. Technical Publication 2280, Metals Technology, Sept., 1947. The American Institute of Mining and Metallurgical Engineers, Inc., New York.

4. Discontinuous grain growth ---- Grain growth in inhibited materials is the sudden development of extremly large grain at the coarsening temperature and results duplex structure .
5. Isothermal grain growth -- For a given annealing temperature the grain size at first increase with annealing temperature at high rate, but then the rate of growth gradually decrease.

The summary of Jeffries and Archer(4) states that grain growth always follows the laws listed below:

1. The grain size of cast metals is usually smaller the more rapid the rate of solidification.
2. The grain size in cast metals which undergo no phase change in the solid state cannot be changed appreciably by heating below the melting point.
3. Grain growth in the solid state may occur in:
   (a) Worked metals
   (b) Compressed powders
   (c) Electrodeposited metals
   (d) In some metals when they change allotropic form
   (e) In alloys in which a new phase is formed in the solid state.

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(4) Jeffries, Zay and Robert S. Archer, The Science of Metals, McGraw-Hill Book Co., New York, pp. 117 - 118.

4. The recrystallization temperature is lower:
    (a) The greater the amount of deformation
    (b) The smaller the grains prior to deformation
    (c) The purer the metal
    (d) The lower the temperature at which the deformation is effected
    (e) The longer the time of heating.
5. In any given metal the grain size, after complete recrystallization, is normally smaller the lower the recrystallization temperature.
6. Above the recrystallization temperature the grain size is normally greater the higher the temperature and the longer the exposure.
7. The higher the temperature the more rapid the grain growth.
8. Certain conditions of non-uniformity sometimes give rise to the formation of abnormally large grains. This is called germination .
9. The non-uniformities giving rise to germination are grain size contrast, strain gradients, temperature gradients, concentration gradients, and obstruction gradients.
10. The presence of foreign matter or a second phase introduces mechanical obstruction to grain growth.
11. The presence of a certain amount of obstruction matter promotes germination.
12. The higher the germinative temperature the larger will be the grains and the quicker will they form.

13. The more rapid the heating through the germinative temperature range, at least within limits, the finer will be the grain size.

Carpenter and Elam(5) studied the grain growth phenomenon and concluded that:

1. Growth may take place either of a large crystal into a smaller one, or of a small into a larger, i.e. crystal growth does not depend upon the relative size of the crystals.
2. The relative orientation of the crystal which is being absorbed and that which is growing does not appear to be related.
3. A crystal which is itself being invaded by one crystal may grow at the same time at the expense of another.
4. The rate of growth is not constant for any given time at any particular temperature.
5. The change of orientation is accompanied by a difference of level of the surface which is the boundary marking observed.

3. Theories of Grain Growth

A theory suggested by Ewing and Rosenhain(6) was based on the assumption that even in the purest metals the crystals are surrounded by a eutectic layer. They considered that "grain growth is due to

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(5) Carpenter, H.C.H. and C. F. Elam, Crystal Growth and Recrystallization. Journal of Institute of Metals, Vol. 24, No. 2, 1920, pp. 83 - 131.

(6) See Footnote (5), p. 85.

"solution and diffusion" of the pure metal constituting the crystals into this fusible and mobile eutectic, forming the intercrystalline cement. This diffusion results in the growth of one crystal at the expense of the other, owing to difference in solubility of the crystal faces on the either side of the eutectic film, and it seems probable that this phenomenon of directed diffusion is really a form of electrolysis."

Rosenhain(7) explained that the molecules in the intercrystalline layer are continually trying to adopt a crystalline arrangement in conformity with that of one or other of the adjacent crystals. The effect of heat would be to cause an increased vibration in the crystal as well as in the amorphous cement, and would cause a molecule from the crystal to join the amorphous layer without any neucleation being required.

Jeffries(8) considered that when the temperature is such that crystal growth occurs "an atom of amorphous material" will be forced into regular arrangement, owing to its greater vibrational energy.

For the question of "why the absence of growth in cast metals without deformation", Chappell(9) gave two possible explanations of

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(7) Rosenhain, Walter, Intercrystalline Cohesion of Metals, Journal of Institute of Metals, Vol. X, No. 2, 1913, p. 134.

(8) Jeffries, Zay, The Science of Metals, McGraw-Hill Book Co., New York, p. 143.

(9) Carpenter, H.C.H. and C. F. Elam, Crystal Growth and Recrystallization, Journal of Institute of Metals, Vol. 24, No. 2, 1920, p. 93

the rule of plastic deformation in promoting the union of ferrite grains as:

".1. If similar orientation is the only condition necessary for union of adjacent crystals, then plastic deformation may be considered to act by increasing the potential energy of the crystals, and thus enabling them to bring about the rotation of their axes parallel to each other with greater ease and rapidity on rise of temperature."

".2. The union of ferrite crystals may be considered analogous to the welding together of two pieces of iron, in which work is necessary to break up any separating layer of oxide or flux and thus establishing molecular contact between metallic surfaces."

Grain growth in the solid state takes place by the gradual absorption of some of the grains by others. Howe(10) describes that "grains grow by coalescence" when the whole grains gradually changed orientation until two adjacent ones had the same orientation, after which they suddenly merged". But according to Rosenhain and Carpenter and Elam's(11) results, the grain growth takes place always by "boundary migration" not by a suddenly complete 'colescence' between two crystals.

Equilibrium

Carpenter(12) showed that in the case of aluminum annealed for four months at 550°C no appreciable increase in grain size resulted after three weeks at that temperature. It was considered probable that equilibrium had been reached after this period of time.

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(10) Howe, Henry M., Transactions of the American Institute of Mining Engineers, Vol. LVI, pp. 582 - 599.

(11) Carpenter, H.C.H. and C. F. Elam, Crystal Growth and Recrystallisation, Journal of Institute of Metals, Vol. 24, No. 2, 1920, pp. 83 - 131.

B. Production of single crystals

From the literature, it can be shown that there are four methods of producing metallic single crystals.

Group 1. From the liquid state,

Group 2. From the gaseous state,

Group 3. From the electrolysis of a liquid, and

Group 4. By recrystallization in the solid state.

Group 1:

(a) Tammann's Method(13) —— Tammann succeeded in making Bi single crystals of a maximum length of 20 cm. by under cooling the molten metal 0.1 to 0.3°C below its melting point in glass tubes of a maximum diameter of 1.5 mm. In the closed end of the tube a crystal nuclei was formed which absorbed the remaining liquid metal in the tube and thus formed a single crystal.

Obreimow and Schubnikow(14) using this method obtained the single crystals of V, Zn, Sn, Sb, Mg, Al and Cu.

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(12) Carpenter, H.C.H. and C. F. Elam, Crystal Growth and Recrystallization, Journal of Institute of Metals, Vol. 24, No. 2, 1920, p. 125.

+(13) Tammann, Lehrbuch der Metallographic (1923) p. 16.

+(14) Obreimow and Schubnikow, Zeitschr. fur Physik, 1924, Vol. 25, p. 31.

(b) **Czochralski's Method**(15) —— Czochralski started with a crucible containing the molten metal at the freezing temperature. This metal was drawn upward from the crucible by means of an auxiliary wire, which moved vertically with a uniform motion. Some distance above the surface of the molten metal solidification started and if care was taken to keep the speed with which the auxiliary wire drawn upward the same as the speed of crystallization, long single crystal wires could be made. The wire was withdrawn not too quickly and not too slowly. The rate of the crystallization was about 0.2 mm. per second.

This method has been extended and improved by Gompers(16) and Mark Polanyi and Schmid(17), who made single crystals of Pb, Sn, Al, Bi, and Cd.

Group 2:

These methods start with a small single crystal, obtained according to another method. This single crystal serves as a nucleus and grows to a larger single crystal.

a. **Ledyguine's Method**(18) —— In this method a mixture of tungsten

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+(15) Czochralski, Zeitschr. fur Physik, 1918, Vol. 92, p. 219

+(16) Zeitschr. fur Physik, 1922, Vol. 8, p. 184.

+(17) Zeitschr. fur Physik, 1923, Vol. 12, p. 60.

+(18) U.S. Patent No. 575002 and No. 575668, 1893.

chloride vapor (WCl_6) and H_2 strikes a glowing tungsten wire; another similar scheme was that of Iseki(19), in which WCl_6 vapor alone strikes the glowing tungsten wire.

b. Koref's method(20) —— The above reaction mentioned was used by Koref to make a glowing tungsten single crystal wire grow. The wire is held at about 1000°C. Hydrogen at a pressure of 12 mm. of Hg passes through a reservoir which leads into the furnace. The original wire grows as a single crystal. Koref and Fischvoigt(21) have stated that they succeeded in getting single crystals of Mo, Ta, Fe, Zr, and Ti in a similar way.

c. Van Arkel's Method(22) —— In this case, a tungsten wire hanging vertically in pure WCl_6 vapor was made into a thick tungsten bar of a maximum thickness of 9 mm. at a temperature 1600°-1800°C. The chlorine arising from the decomposition is again converted into WCl_6 by tungsten powder located at the bottom of the reaction

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+(19) English Patent No. 3509-C, 1906

+(20) Koref, Zeitschr. fur Elektr. 1922, Vol. 28, p. 511.

+(21) Koref, Zeitschr. fur techn. Physik, 1925, p. 296.

+(22) Van Arkel, Physica, Vol. 2, 1922, p. 56.

vessel. The powder is held at 400°C. With this method single crystals of molybdenum also were produced(23).

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*(23) Geiss and V. Liempt, Zeitschr. fur Metallk (Aug., 1924)

Group 3 - Method of J. A. M. Van Liempt(24)

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*(24) J. A. M. Van Liempt, Zeitschr. fur Elektr., 1925, p. 249.

In this method, the single crystal was produced electrolytically at the surface of the metal, the latter being used as a cathode. In this way a much thicker single crystal was obtained from a small single crystal wire, which served as a cathode in a bath of molten sodium tungsten. The cathode wire was placed in the axis of a hollow tungsten cylinder which served as an anode.

Group 4:

a. Method of Schaller and Orbig(25) —— This method was worked out by Schaller and Orbig. It is a modification or adaptation of making pressed or extruded tungsten filaments. In this case very finely divided tungsten powder was made into a paste with a binder and was

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*(25) German Patent 291994, 1913. Bettger, Zeitschr. fur Elektr. 1917, Vol. 23, p. 121.

"squirted" into thin threads through diamond dies. Then these filaments were heated at a high temperature by means of electric current. During this process sintering takes place and the small tungsten crystals grow into larger crystals. The extruded and dried wire was passed through a region of very high temperature (2500°C) with a speed equal to the rate of grain growth (up to 0.1 cm./sec.)

Characteristic of the process is the fact that its success is promoted by adding a small percentage of thoria (2%) to the tungsten. ThO_2 is a highly refractory oxide, which does not even posses an appreciable vapor pressure at 2500°C. The part it plays in the formation of single crystals is not known with certainty. Drawn tungsten wire has also been made into a single crystal wire by heating in a manner similar to that used in the Pintsch(26) process.

b. Sauveur's method(27) ---- In 1912, Sauveur was the first to use the method of critical strain for the production of very large iron crystals. Later, Czochralski, Davey, Carpenter and Elam(28) successfully applied the same principle to various metals.

+(26) Goucher, English Patent 174714, (1920).

+(27) Sauveur, Metallography and Heat Treatment of Iron and Steel, v. 265.

+ Note: From Footnote (13) to (27), also see J. A. M. Van Liempt, The Production of Metallic Single Crystals, A.I.M.E. Proceedings of The Institute of Metals Division, 1928, pp. 307 - 315.

(28) Carpenter, H.C.H. and C.F. Elam, Crystal Growth and Recrystallization, Jour. of Institute of Metals, Vol. 24, No. 2, 1920, pp. 83-131

C. Principles of Powder Metallurgy As Applied to Single Crystals

1. Compression:

In the field of powder metallurgy, the processes used are pressing and sintering, so for the purpose of maximum production, such factors as the maximum pressure, the rate of pressure, the maximum temperature of sintering, the time of sintering, the atmosphere of sintering, the rate of heating and cooling, and the purity of powder used, size of powder, should be controlled.

The purpose of the compacting process is the shaping of metal powders to compacts with sufficient coherence to permit the transfer to the sintering furnace, and with characteristics insuring satisfactory performance during sintering as well as the desired properties of the final sintered product.

If the part is relatively thin and of uniform height, the powder may be compacted by "single" action that is by compression from one side only, otherwise, "double" action should be used.

By the single action, compacting, the compact thus produced will have a higher density in its upper parts. In the case of double action, high pressure powder density will be built up in the vicinity of both punches, and minimum density will be exhibited in intermediate portions of the compacts.

The application of pressure decreases the porosity and at the same time increases the cohesion between particles by increasing the number of points of contact, as well as by causing particles of irregular shapes to interlock.

In general, the density of a compact increases mostly in the low and medium pressure range, The maximum green density attainable by cold pressing depends not only upon the applied pressure but also upon the rate of pressure application.

The rate of pressing is limited by several factors. The most important among these is the effect of compression speed on the amount of air entrapped during compaction.

A slow pressure rate also reduces the friction losses and increases the uniformity of pressure distribution, and of density throughout the compact. This is particularly true with powders having a tendency for bridge formation and in case of compact designs characterized by relative high thickness in the direction of compression.

Because of the fact that the plastic deformability of compacts can be increased with increasing temperature, so, in industry using, hot pressing is employed to increase the density, hardness and tensile strength of compacts.

2. Sintering:

According to Oxford New English Dictionary, 'Sintering' means "to adhere in a mass by partial fusion". The main purpose of the sintering process of powder metallurgy is the transformation of the "green" compact, consisting of individual particles, to a mass in in which the particles have lost their identity, and recrystallization and grain growth have taken place across the former particle boundaries.

Schwarzkopf(29) concludes that the conditions which can be

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(29) Schwarzkopf, Paul, Powder Metallurgy. MacMillan Co., 1947, p. 82.

controlled during the sintering operation are:

(1) The sintering atmosphere

(2) The rate of heating

(3) The maximum temperature of sintering

(4) The time period kept

(5) The rate of cooling.

In general, the atmosphere during sintering should be controlled. The purpose of atmosphere control is as follows:

a. It must prevent undesired reactions, such as oxidation, carburization, and decarburization.

b. It must perform desired reactions, such as the reduction of surface oxides and the removal or replacement of absorbed gases.

To achieve the first of these objects, the atmosphere must be free of certain components, or at least free of excessive amounts of certain components, such as oxygen, moisture, sulfur dioxide, and hydrogen sulfide. The second object, on the other hand, requires the presence of certain generally reducing components, such as hydrogen or carbon monoxide.

The effect of sintering temperatures depends on the pressure applied during the compacting operation. Powders pressed at extremely high pressure frequently show a decrease of density with increasing sintering temperature due to the expansion of the compact because of the escape of gases.

In general, it is possible to obtain similar effects with short sintering periods at high temperatures and with long sintering periods at low temperatures. For economical reasons, it is in most cases preferable to work at the highest possible sintering temperature, in order to reduce the processing time to a minimum.

Recrystallization and grain growth of powder compacts

The close relationship between sintering and the crystallization process in metal makes it necessary to base a theory of powder metallurgy on definite concepts regarding the mechanism of processes such as recrystallization and grain growth.

In Sauerwald's(30) first publication, he found that the temperature of beginning of grain growth is independent to the compacting pressure and to be characteristic for each metal. But, later on, Sauerwald maintains that recrystallization processes can take place in sintered metals without previous plastic deformation. This assumption would indicate a principal difference in sintered and reguline metals. In his latest paper, Sauerwald distinguishes between different ranges of compacting pressures. At low pressures, only elastic deformations are assumed to take place; at medium pressures, plastic deformation and elastic deformation both take place, but plastic deformation only effects on the surface; at high pressure, plastic deformations involves the whole particle.

From the works of Smithells, Pitkin, and Avery(31), who publish

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(30) Schwarzkopf, Paul, Powder Metallurgy. MacMillan Co., 1947, p. 273

(31) Schwarzkopf's Powder Metallurgy, MacMillan Co., 1947, p. 275.

the results of measurements of dimensional changes and of changes of the watt-temperature relationship during the sintering of tungsten, it was concluded that grain growth begins at 1,227°C (2,240°F) with compacts pressed at 8 tsi.; and at 927°C (1,700°F) with pressures of 32 tsi. This conclusion was in direct opposition with the results of Sauerwald.

III

Copper Powders Used in Investigation+

1. Type: Copper powder C type 200 mesh, from Charles Hardy, Inc. New York City, N. Y. Copper content - 99.00%.

2. Size analysis:

Mesh	Per Cent
100	0
100 - 150	0
150 - 200	0.1 - 1.0
200 - 250	0.2 - 10.0
250 - 325	2.0 - 10.0
-325	90.0 - 97.0

3. Tap density of copper powder: 2.94 g/cc.

4. Loss of weight in hydrogen: 0.15 - 0.70%

+ Data supplied by Company

IV

Grain Size Determination

For the study of grain size, the ASTM standard grain size eye piece was used. The true size of each scale was calculated as follows:

No. of grain size in scale (100x)	No. of grains per sq. inch under 100x	Area of each grain in sq. in. of 100x
1	1	1/100
2	2	1/200
3	4	1/400
4	8	1/800
5	16	1/1600
6	32	1/3200
7	64	1/6400
8	128	1/12800

The area (A) of each grain in sq. in. under different magnification is calculated by the following formula:

A = 1/ (magnification x No. of grains per sq. in.)

For a small area, there will be no great difference in linear dimension in assuming that it is a circle or a square. For simplicity, the linear dimension of each grain for each different grain size number at different magnification is calculated and listed on the next page assuming that all grains are square.

Table 1

Linear Dimension of Grains
For different grain size
numbers at different magnifications
(inch x 10^{-2})

No. of Scale	450x	100x	50x	40x	25x
1	4.67	10.00	14.20	14.80	20.00
2	3.32	7.05	10.00	10.42	14.20
3	2.34	5.00	7.05	7.45	10.00
4	1.65	3.55	5.00	5.25	7.05
5	1.17	2.50	3.55	3.70	5.00
6	0.83	1.76	2.50	2.62	3.55
7	0.58	1.25	1.76	1.85	2.50
8	0.41	0.81	1.25	1.32	1.76

V

Equipment

I. For compression:

A. Riehle Bros. Compressing Machine ————

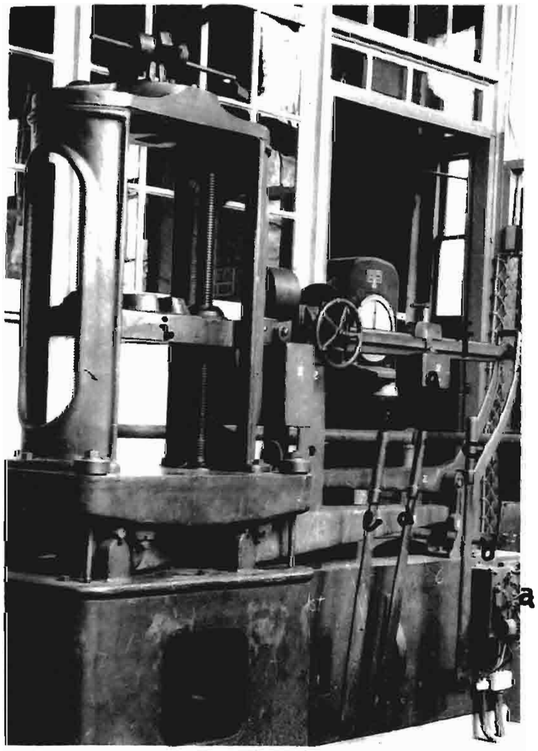
This machine can be used for either tension or compression with the load range from 1000 to 50,000 lbs. It has four different speeds, which are controlled by different gears. The speeds and the maximum load for each speed are listed in table 2.

Table 2

No. of speed	Max. load (lbs.)	Head Speed (in./min.)
1	10,000	7
2	25,000	3
3	36,000	0.3
4	50,000	0.067

Fig. 1

Riehle Bros. Compression Machine



a — current switch

b — motor switch

c — gear switch

d, e — speed adjustment

f — load lever

g — load adjustment

h — screw column

i — press plate

B. Dies used:

1. Single Action Round Die ---- The single action round die is made by hardened alloy steel. Its dimensions are listed as follows:

Height	3.0625 in.
Outside diameter	1.0750 in.
Inside diameter	1.0320 in.

The plunge is 1.015 in. in diameter and 0.82 sq. in. in cross section area.

Fig. 2 Single Action Die

2. Double action die* ---- It is also called floating die. In construction, the die assembly is supported by the base. When the upper punch advances, friction between powder and walls or mechanical action, forces the die assembly downward over the lower punch, thus compressing the springs. This compression produces a movement of the die walls relative to the stationary lower punch, so that compression from both sides (double action) results from the movement of only one punch.

* Supplied through the courtesy of Charles Hardy, Inc., New York, N. Y.

Fig. 3 Double action die

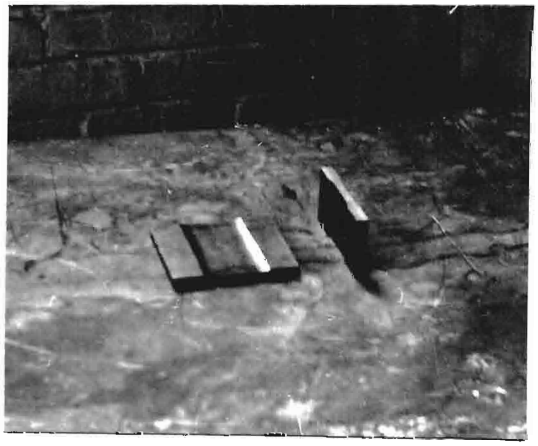


Fig. 4 Punch of double action die

II. For Sintering and Annealing:

Electric Multiple Unit Furnace ——— This furnace is an electric resistance furnace with automatic temperature control and rate of heating or cooling adjustment. It uses 230 volts A.C. current and 3600 watts, its maximum temperature is 1850°F (1010°C).

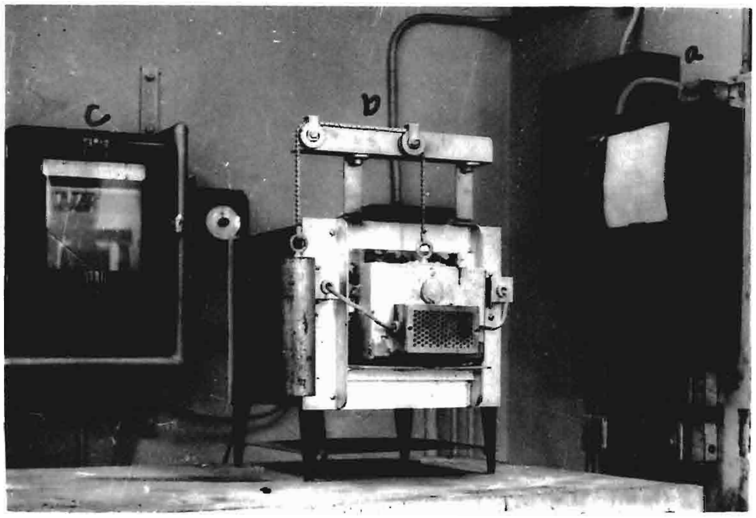


Fig. 5 Electric Multiple Unit Furnace

a —— Main switch of current

b —— Furnace

c —— Automatic controlling and record

2. **Cast Iron Box** ——— It was used for sintering and annealing the specimens. The specimen was covered with graphite powder and the box was sealed with cement so that a reducing atmosphere was produced inside the box.

Fig. 6 Cast Iron Box

III. Equipment Used for Deformation:

1. Carver Laboratory Press ----

This machine is operated by an oil ram. The maximum pressure is 16,000 lbs. It can be used for hot pressing by using 110 volts electricity. In this work, it was used to deform the flat specimens by cold compression.

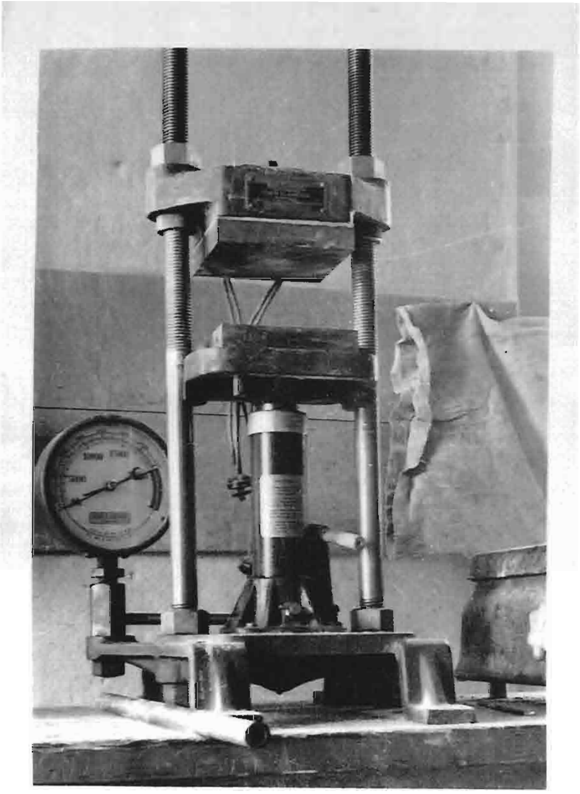


Fig. 7. Carver Laboratory Press

2. Dillon Tensile Testing Machine -----

It is used only for tensile testing of wire. The maximum load which can be used is 100 pounds. In this research, it was used to elongate the copper wire.

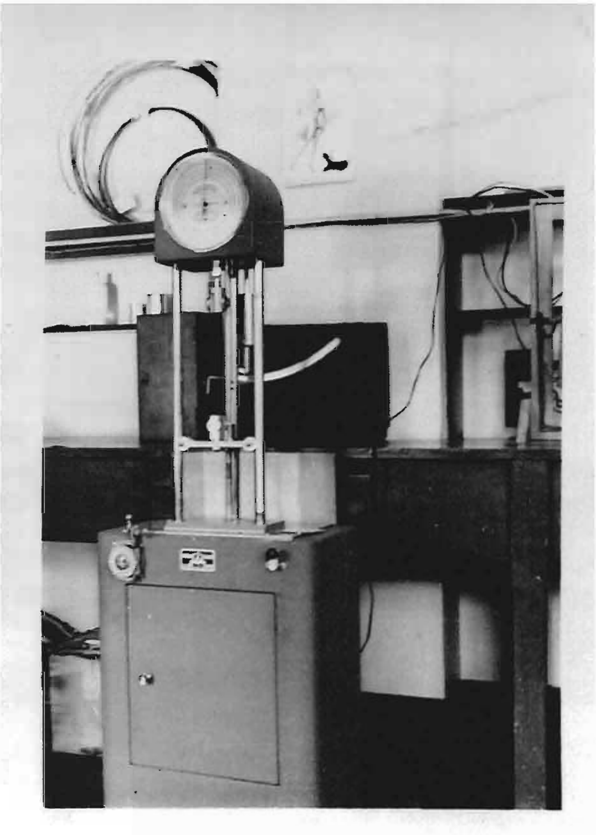


Figure 8. Dillon Tensile Testing Machine

3. Southwark Stress-Strain Testing Machine -----

It can be used either for tension or for compression. It is operated by compressed air from the pump. Both the load and the rate of running can be adjusted. The maximum load, which can be used is 20,000 lbs. There are three speeds which can be selected:

High speed	1,000 to 20,000 lbs./min.
Medium speed	250 to 5,000 lbs./min.
Low speed	50 to 1,000 lbs./min.

For this work, it was used to deform the compressed tensile bars by tension, and the low speed was used.

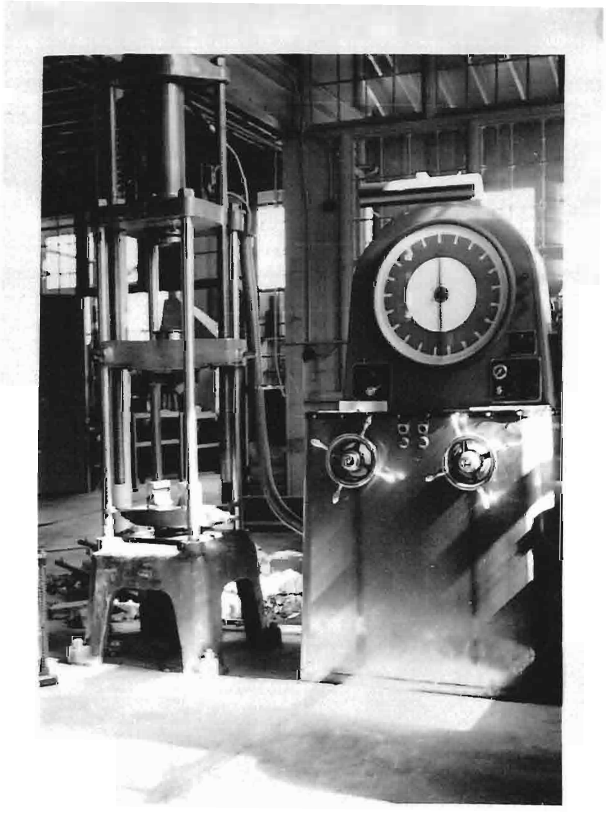


Fig. 9 Southwark Stress-Strain
Testing Machine

VI

Experimental Procedure and Resulting Data

Experiment 1. Repeated deformation and annealing of compressed powder flats

By using the single action small round die, 38 small flats were prepared. They were pressed by a series of pressure from 2.43 tsi to 30.5 tsi (max. pressure on the pressing machine) in different groups. The pressure 2.43 tsi is the lowest which can be used; below that, the samples cannot be ejected safely after compressing.

For sintering, the different temperature and time were used for different groups. (see data in the attached table) All compacted specimens were put in alloy cast iron boxes and covered with graphite powder and the boxes were sealed with cement so that a reducing atmosphere was produced to protest the specimens from oxidation when heated. The boxes were heated in an electric Multiple Unit Furnace with automatic temperature control. The heating rate was about 150°C per hour to fixed temperature and the boxes were cooled in the furnace with the rate about 100°C per hour.

After sintering, each group of specimens was deformed by cold working with the Carver Laboratory Press (Fig. 7) to secure the critical deformation. The amount of deformation was not easy to control, so the results may vary somewhat from idea requirements. The deformation was calculated from the reduction of thickness of the samples. For convenience of the grain size study, the samples were cut to small squares about a half an inch on each side. Then, the cold deformed specimens were annealed in the same electric furnace that was used for the sintering operation.

The annealed specimens then were cold deformed again and followed by annealing. The deformation, annealing cycle was repeated eight times. The data for compressing, sintering, cold-deformation-annealing, and the grain size after sintering and annealing are listed in Tables 3, 4, 5, and 6 respectively.

Unfortunately, all specimens were melted in the ninth annealing cycle due to the failure of the temperature controlling device, therefore, they could not be photographed.

Data and Results

Table 3 Compressing

No. of Sample	Pressure tsi.	Speed Of Pressing	Thickness Of Compace (inch)	Volume of Compact (Cu. in.)	Density g/cc	Wt. of Compact (g)
1	2.43	4	0.320	0.262	4.58	19.70
2	3.65	4	0.312	0.255	4.76	19.90
3	4.86	4	0.301	0.246	4.92	19.85
4	7.30	4	0.266	0.208	5.85	19.90
5	9.75	4	0.246	0.202	6.00	19.80
6	11.60	4	0.248	0.203	6.02	19.95
7	12.20	4	0.242	0.199	6.08	19.85
8	12.80	4	0.237	0.194	6.30	19.90
9	13.40	4	0.234	0.190	6.37	19.90
10	14.00	4	0.231	0.189	6.45	19.80
11	14.60	4	0.235	0.192	6.53	19.90
12	15.20	4	0.223	0.183	6.61	19.85
13	15.80	4	0.229	0.187	6.55	19.95
14	15.70	4	0.215	0.176	6.81	19.80
15	17.10	3	0.221	0.182	6.67	19.85
16	17.70	4	0.218	0.179	6.78	19.90
17	18.30	3	0.222	0.182	6.67	19.85
18	18.30	4	0.199	0.162	7.58	19.90
19	24.20	4	0.204	0.167	7.30	19.80
20	24.20	3	0.205	0.168	7.29	19.80
21	24.20	3	0.205	0.168	7.29	19.80
22	25.00	3	0.201	0.165	7.35	19.90
23	25.60	4	0.198	0.162	7.46	19.92
24	26.30	4	0.197	0.161	7.50	19.90
25	27.20	4	0.197	0.161	7.60	19.93
26	28.10	4	0.198	0.162	7.46	19.90
27	28.60	4	0.196	0.160	7.62	19.93
28	29.20	4	0.203	0.166	7.32	19.85
29	29.20	4	0.198	0.162	7.50	19.90
30	29.20	4	0.198	0.162	7.50	19.91
31	29.20	4	0.197	0.161	7.60	19.89
32	29.80	4	0.198	0.162	7.46	19.88
33	30.50	4	0.196	0.160	7.62	19.90
34	30.50	4	0.198	0.162	7.53	19.93
35	30.50	4	0.198	0.162	7.55	19.95
36	30.50	4	0.197	0.161	7.60	19.90
37	30.50	4	0.198	0.162	7.55	19.96
38	30.50	4	0.197	0.161	7.60	19.90

\+ All specimens are pressed by use of 20 g. powder and with same single action round die with diameter 1.035 inches and cross section area as 0.822 sq. in.

Table 4 Sintering

No. of Sample	Sintering Temp. (°F)	Sintering Time (hr.)	Thickness (in.)	Diameter (in.)	Volume (cu.in.)	Weight (g)	Density (g/cc)
1	1700	17	0.230	0.840	0.127	18.65	9.05
2	1700	17	0.248	0.866	0.145	18.80	8.00
3	1700	17	0.257	0.902	0.164	18.90	7.50
4	1700	17	0.221	0.896	0.139	19.00	8.35
5	1700	17	0.212	0.923	0.141	18.00	8.10
6	1800	3	0.224	0.918	0.140	19.00	8.30
7	1800	3	0.213	0.924	0.146	18.80	7.90
8	1800	3	0.212	0.941	0.147	18.70	7.80
9	1800	3	0.218	0.946	0.155	18.75	7.40
10	1800	3	0.209	0.948	0.148	18.85	7.80
11	1800	3	0.207	0.947	0.144	18.80	8.00
12	1800	3	0.216	0.965	0.158	18.85	7.35
13	1800	3	0.214	0.972	0.159	18.84	7.30
14	1800	3	0.213	0.970	0.158	18.86	7.36
15	1800	3	0.216	0.980	0.163	18.79	7.10
16	1800	3	0.214	0.992	0.164	18.82	7.06
17	1700	1	0.202	0.957	0.155	18.92	7.45
18	1700	1	0.205	1.027	0.170	18.88	6.70
19	1700	1	0.204	1.010	0.164	18.90	7.05
20	1700	1	0.206	1.013	0.166	18.87	7.00
21	1700	1	0.213	1.012	0.164	18.88	7.06
22	1700	1	0.203	1.026	0.177	18.90	6.55
23	1700	1	0.205	1.022	0.168	19.00	6.95
24	1700	1	0.209	1.036	0.177	18.92	6.80
25	1700	1	0.217	1.034	0.182	18.87	6.36
26	1700	1	0.204	1.023	0.169	18.88	6.88
27	1700	1	0.209	1.031	0.179	18.89	6.52
28	1700	1	0.214	1.028	0.179	18.85	6.46
29	1700	1	0.211	1.039	0.176	18.85	6.55
30	1800	3	0.220	1.038	0.183	18.90	6.30
31	1800	3	0.206	1.040	0.176	18.87	6.57
32	1800	3	0.213	1.040	0.182	18.85	6.37
33	1800	3	0.214	1.044	0.184	18.80	6.30
34	1700	1	0.208	1.031	0.176	18.79	6.52
35	1700	1	0.206	1.038	0.177	18.84	6.53
36	1700	1	0.205	1.033	0.172	18.90	6.75
37	1700	1	0.208	1.034	0.176	18.90	6.58
38	1700	1	0.215	1.038	0.182	19.00	6.40

Table 5

Deformation and Annealing

No. of Sample	I Annealed			II Annealed			III Annealed		
	Def. (%)	Temp. (°F)	Time (hr.)	Def. (%)	Temp. (°F)	Time (hr)	Def. (%)	Temp. (°F)	Time (hr)
1	9.8	1600	40	3.0	1600	48	6.0	1600	60
2	8.3	1600	40	5.0	1600	48	8.0	1600	60
3	5.1	1600	40	4.0	1600	48	7.0	1600	60
4	5.0	1600	40	6.0	1600	48	6.5	1600	60
5	5.1	1600	40	3.0	1600	48	5.0	1600	60
6	4.5	1600	64	7.0	1600	40	3.5	1600	48
7	6.3	1600	64	2.6	1600	40	3.5	1600	48
8	6.3	1600	64	5.9	1600	40	2.3	1600	48
9	7.0	1600	64	3.8	1600	40	5.7	1600	48
10	11.4	1600	64	3.7	1600	40	2.8	1600	48
11	4.1	1600	64	4.2	1600	40	2.0	1600	48
12	2.8	1600	64	7.2	1600	40	9.0	1600	48
13	7.4	1600	64	5.5	1600	40	3.8	1600	48
14	5.6	1600	64	7.0	1600	40	8.5	1600	48
15	5.6	1600	64	9.5	1600	40	6.0	1600	48
16	5.6	1600	64	4.6	1600	40	19.0	1600	48
17	9.5	1600	64	2.0	1600	40	8.0	1600	48
18	10.0	1600	64	3.5	1600	40	9.0	1600	48
19	4.6	1600	64	3.0	1600	40	6.2	1600	48
20	6.0	1600	64	3.0	1600	40	18.0	1600	48
21	4.7	1600	64	6.7	1600	40	3.2	1600	48
22	5.4	1600	64	1.3	1600	40	2.5	1600	48
23	7.4	1600	64	1.8	1600	40	6.2	1600	48
24	12.0	1600	64	5.4	1600	40	4.0	1600	48
25	5.9	1600	64	5.3	1600	40	3.2	1600	48
26	8.1	1600	64	3.3	1600	40	3.2	1600	48
27	14.0	1600	64	4.4	1600	40	3.2	1600	48
28	17.4	1600	64	5.0	1600	40	4.2	1600	48
29	11.6	1600	64	4.1	1600	40	5.0	1600	48
30	26.7	1600	64	12.0	1600	40	4.0	1600	48
31	14.8	1600	64	4.5	1600	40	15.0	1600	48
32	14.7	1600	64	11.4	1600	40	6.0	1600	48
33	15.3	1600	64	10.8	1600	40	4.5	1600	48
34	9.5	1600	64	3.1	1600	40	4.3	1600	48
35	12.5	1600	64	5.7	1600	40	6.7	1600	48
36	12.9	1600	64	9.3	1600	40	8.5	1600	48
37	21.2	1600	64	4.3	1600	40	5.0	1600	48
38	14.7	1600	64	2.5	1600	40	1.7	1600	48

Table 5 Deformation and Annealing (Cont.)

No. Of Sample	IV Def. (%)	Annealed Temp. (°F)	Time (hr)	V Def. (%)	Annealed Temp. (°F)	Time (hr)	VI Def. (%)	Annealed Temp. (°F)	Time (hr)
1	5.6	1600	105	5.0	1700	63	6.0	1700	48
2	5.0	1600	105	5.5	1700	63	6.2	1700	48
3	4.4	1600	105	5.0	1700	63	4.0	1700	48
4	4.6	1600	105	7.5	1700	63	5.0	1700	48
5	3.8	1600	105	4.0	1700	63	4.1	1700	48
6	9.5	1600	60	3.7	1600	105	5.0	1700	63
7	3.5	1600	60	2.2	1600	105	3.0	1700	63
8	4.2	1600	60	3.8	1600	105	4.0	1700	63
9	3.8	1600	60	4.8	1600	105	4.1	1700	63
10	3.5	1600	60	2.2	1600	105	4.0	1700	63
11	5.5	1600	60	2.2	1600	105	2.5	1700	63
12	2.2	1600	60	2.5	1600	105	3.0	1700	63
13	5.0	1600	60	5.5	1600	105	4.1	1700	63
14	4.8	1600	60	5.0	1600	105	4.2	1700	63
15	3.5	1600	60	1.9	1600	105	4.5	1700	63
16	5.2	1600	60	2.8	1600	105	5.0	1700	63
17	3.0	1600	60	1.6	1600	105	3.3	1700	63
18	3.3	1600	60	4.0	1600	105	2.5	1700	63
19	4.2	1600	60	1.6	1600	105	3.0	1700	63
20	4.0	1600	60	3.5	1600	105	3.5	1700	63
21	3.0	1600	60	1.6	1600	105	1.5	1700	63
22	3.8	1600	60	3.1	1600	105	3.2	1700	63
23	1.5	1600	60	3.5	1600	105	2.5	1700	63
24	4.5	1600	60	3.6	1600	105	2.0	1700	63
25	1.0	1600	60	2.7	1600	105	1.0	1700	63
26	6.8	1600	60	3.7	1600	105	0.8	1700	63
27	3.5	1600	60	5.6	1600	105	0.5	1700	63
28	4.5	1600	60	2.8	1600	105	1.8	1700	63
29	3.0	1600	60	2.0	1600	105	2.0	1700	63
30	6.0	1600	60	2.2	1600	105	2.1	1700	63
31	7.2	1600	60	3.2	1600	105	4.0	1700	63
32	4.0	1600	60	2.2	1600	105	3.2	1700	63
33	3.8	1600	60	2.8	1600	105	3.7	1700	63
34	5.0	1600	60	18.0	1600	105	2.5	1700	63
35	2.0	1600	60	2.8	1600	105	2.0	1700	63
36	3.0	1600	60	3.9	1600	105	5.0	1700	63
37	7.0	1600	60	1.0	1600	105	1.0	1700	63
38	3.0	1600	60	2.7	1600	105	2.0	1700	63

Table 5 Cont.

Deformation and Annealing

No. Of Sample	VII	Annealed		VIII	Annealed	
	Def. (%)	Temp. (°F)	Time (hr)	Def. (%)	Temp. (°F)	Time (hr)
1	6.2	1700	24			
2	4.0	1700	24			
3	5.8	1700	24			
4	6.0	1700	24			
5	3.0	1700	24			
6	4.5	1700	48	6.0	1700	24
7	3.5	1700	48	3.0	1700	24
8	2.2	1700	48	6.0	1700	24
9	4.0	1700	48	4.5	1700	24
10	4.1	1700	48	4.5	1700	24
11	4.5	1700	48	3.0	1700	24
12	3.5	1700	48	5.0	1700	24
13	4.2	1700	48	3.5	1700	24
14	5.5	1700	48	8.0	1700	24
15	2.1	1700	48	4.4	1700	24
16	2.0	1700	48	2.0	1700	24
17	3.0	1700	48	2.2	1700	24
18	3.2	1700	48	3.0	1700	24
19	2.5	1700	48	1.6	1700	24
20	3.6	1700	48	4.0	1700	24
21	1.0	1700	48	2.0	1700	24
22	3.2	1700	48	2.0	1700	24
23	3.5	1700	48	3.8	1700	24
24	7.2	1700	48	4.0	1700	24
25	4.5	1700	48	2.0	1700	24
26	3.0	1700	48	4.0	1700	24
27	5.5	1700	48	3.0	1700	24
28	2.0	1700	48	2.0	1700	24
29	3.1	1700	48	2.0	1700	24
30	3.0	1700	48	4.5	1700	24
31	2.3	1700	48	4.5	1700	24
32	3.3	1700	48	2.5	1700	24
33	3.0	1700	48	4.1	1700	24
34	1.3	1700	48	4.0	1700	24
35	4.0	1700	48	3.0	1700	24
36	3.0	1700	48	5.0	1700	24
37	2.2	1700	48	3.0	1700	24
38	3.0	1700	48	2.0	1700	24

Table 6 Grain Size

No. of sample	After Sintering	I Annealed	II Annealed	III Annealed	IV Annealed		V Annealed		VI Annealed		VII Annealed		VIII Annealed	
	450x	450x	450x	100x	100x	50x	100x	40x	100x	40x	40x	25x	40x	25x
1	5	4	4+	4	3		3+			3	3			
2	4	4+	3	3	3+		2			3	1			
3	3	3+	2	2+	2+			2		1		1		
4	3	3+	2	2	1			1		1+		1+		
5	2	1	1+	1		1		1		1+		1+		
6	3	1	1+	1		1+		1+		1+		1+		1++
7	3	1	1+	1		2		2+		1		1		1+
8	3-	2	1	1-	1			2		1-		1-		1+
9	4	2-	2	2	1			2		2+	1			1
10	4	3	2	2	1			2		2	1			1
11	5	4+	2-	1	1			2		2-	1			1
12	4	3	2	1	1			2-		2+	1			1-
13	5+	4	2-	2-	1-			3		2	1-			1-
14	5	4	2	3+	2			3		3	2		1	
15	5-	5	2-	3	2			3-		3.	2-		1-	
16	6	5-	3	3	3+		2		1		2-		1	
17	6	5	3	4	2		2		2+		3		1-	
18	6-	5	3	5	4		3		2		3-		2	
19	6	5	3	4	3		2		2		3		1	
20	7+	6	3-	4	3-		2		1		4+		2	
21	7	6-	4	3-	3-		2-		2		4		2-	
22	7	6	4	4	3		2		2		4-		2	
23	5	6+	4	4-	4+		3		2-		4		2-	
24	7	6	5	5	4		3		2		4		2	
25	7	6	5	6	5		3		2		4-		2	
26	7	6	5	5	4		3-		2		4		2-	
27	7-	6	5	6	5		4		3		4		3	
28	7	6	6	6	5		4		3		4		3	
29	7	6-	6	6	5		4		3		4-		3	
30	7-	6-	6	6	5		4-		3-		4		3	
31	7+	6	5	5	5		4		3		4		3	
32	7	6	6	6	5		4		3		4-		3-	
33	7	6	5	6	6		4-		3		4-		3-	
34	8	7	6	7	6		5		4		5		4	
35	8	7	6	7	6		5		4		5		4	
36	8	7	6	7	7		5		3		4		3-	
37	8	7	5	7	7		5		4		5		4	
38	8	6	6	7	7		4		3		4		4	

\- means smaller + means larger

All numbers show ASTM grain size scale numbers

Experiment 2. Copper Wire

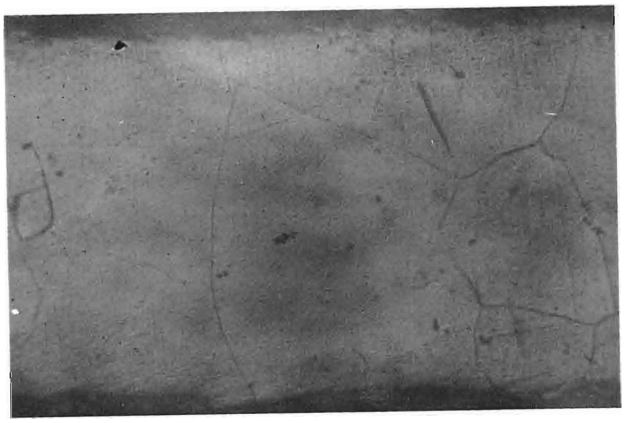
For the purpose of finding the favorable temperature of recrystallization and grain growth of copper, a copper wire with 1/16 inch in diameter was used.

First, due to the fact that most of the wires are prepared by cold drawing, the original deformation is very large and the grain is very small. For stress relief, two specimens were cut from the wire. One was annealed at 650°C for one hour; and the other was annealed at 950°C for two hours and both with slow heating and slow cooling. The latter gave larger grains and showed that the stress was relieved completely.

Second, the annealed copper wire sample (950°C - 2 hours.) was given 2% elongation by using the Dillon Tensile Testing Machine. Then, three samples were cut from this specimen: one was annealed at 920°C for four hours, and another at 950°C for four hours; the third sample at 950°C for five hours. The result of the last one shows the largest grain almost the same as the diameter of the wire (1/16 inch) (Fig. 10).

All of the specimens were put in the alloy cast iron boxes and covered with graphite powder and the boxes were sealed with cement. The cast iron boxes were heated in the electric multiple heating furnace with the same rate of heating and cooling as that used in experiment 1.

Fig. 18



25 X
Copper electric wire with 1/16 inch in diameter was annealed at 950°C for 2 hours, 2% elongation for deformation and annealed at 950°C for 5 hours.

Experiment 3. Compressed Copper Powder Tensile Bars

From the data obtained from Experiment 1, the optimum compressing pressure range for grain growth of copper powder compacts appeared to be from 8 to 12 tons/sq. in. Therefore, in this experiment, nine tensile bars were prepared by compressing copper powder in the double action die and within this pressure range.

After compression, they were sintered at 950°C for five hours by the same furnace and the same method being used to control the atmosphere in the box with graphite powder.

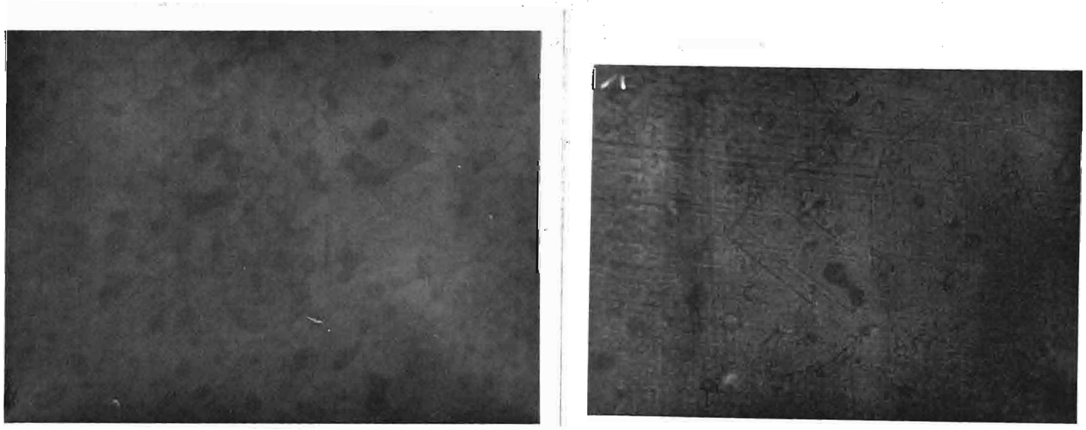
After sintering, the grain size was studied, the specimen using 9 tsi pressure gave a very large crystal as shown in Fig. 11c.

For the elongation of the tensile bars, the Southwark Stress-Strain Testing Machine was used. (Fig. 9). It was determined by the following procedure: at first, mark one inch in length in the uniform cross section part of bars; then the load was applied, until the marked length extends to 1.02 in. Since the gauge used was only approximately rough, the resulting amount of deformation was not as accurate as had been originally desired.

Then, these tensile bars were annealed at 950°C for thirty hours in the same electric furnace by using the method as that used in sintering. Although the grains grew very large as shown in Fig. 12 and 13, unfortunately they were still too small to entirely correct the test piece to a single crystal.

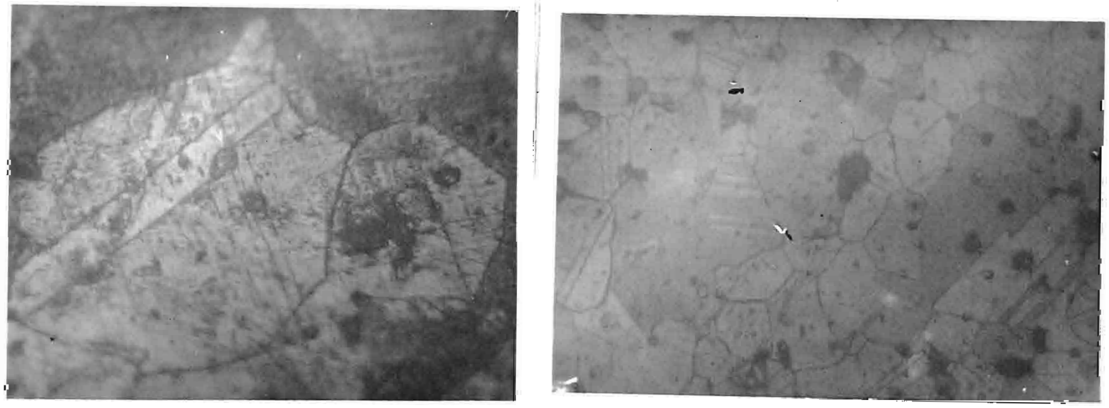
Fig. 11

Grain size of tensile bars
under 250X after sintering
at 950°C (1742°F) for 5 hrs.



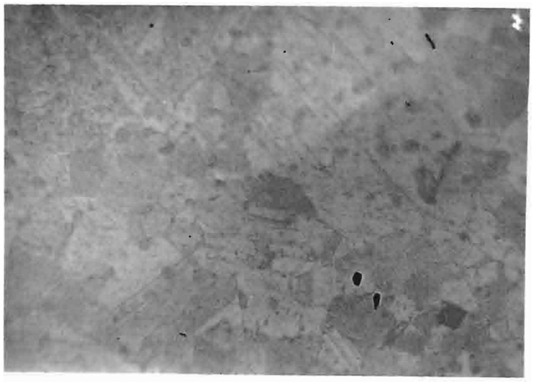
(a) 8 tsi

(b) 9 tsi

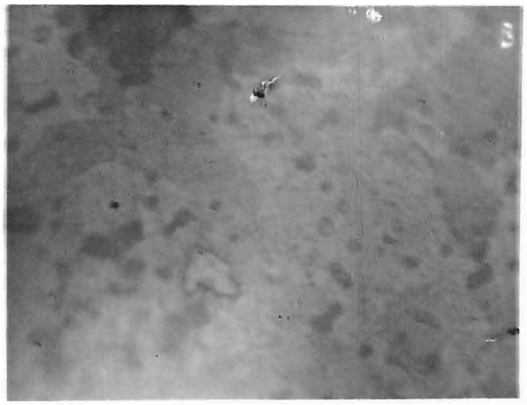


(c) 9 tsi

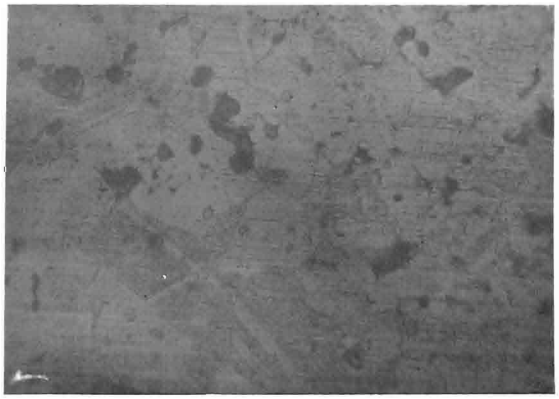
(d) 10 tsi



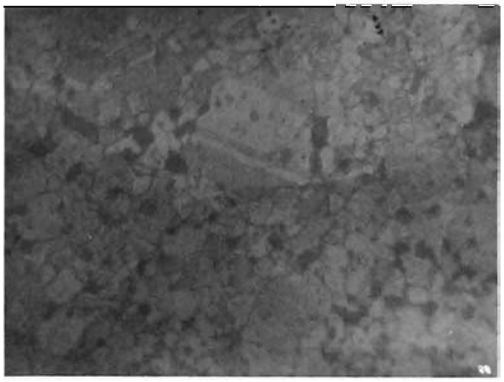
(e) 10 tsi



(f) 11 tsi



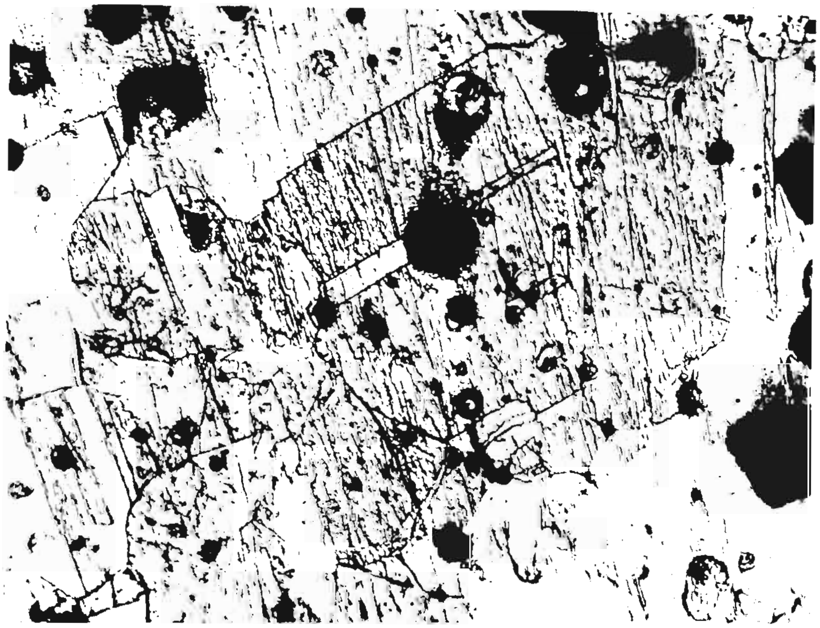
(g) 11 tsi



(h) 12 tsi

Fig. 12

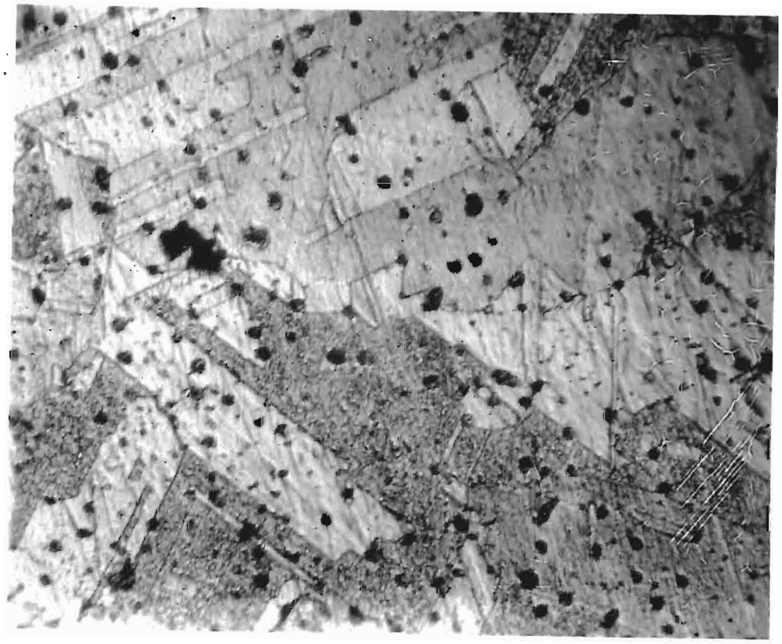
Grain size on the tensile bar
of compressing pressure 9 tsi
sintered at 950°C (1710°F) for
5 hours and 2% elongation for
deformation and annealed at
950°C for 30 hours.



200 X

Fig. 13

Grain size of tensile bar
of 8 tsi compressing pressure
and sintered at 950°C for 5 hours
and 2% elongation for deformation
and annealed at 950°C for 30 hours.



100 X

VII

Discussion of Results

A. Pressing and Sintering

Effect of pressure on grain size ——— From the Experiment 1 of this research, the compressing pressure of from 8 to 12 tons/sq. in. was found to be the optimum range for grain growth. (see Fig. 16)

Due to the very nature of the method used to produce the compacts the grain size was influenced by a number of factors. Some of these had been anticipated and were inherent to the method. Even with slow pressing a stress gradient existed between the surface and center of the specimen. The lack of uniform particle size (commercial powders were used) was reflected in the different size particles having different internal strengths, so when the pressure was applied to the surface of the compacts, the particles of the surface layer tended to move downward filling the pores, thus reducing the distance between each particle and its neighbor. This would tend to increase the density of the compact as a whole would have little, if any, effect on the density of individual particles.

Any pressure greater than that required to fill pore space would tend to deform individual particles. The first particle to be so deformed being there adjacent to the walls and surface of the plunger.

In the work of Experiment 1, most of the specimens showed the larger grains at the position of just under their outer surface. After sintering, the larger grains were located closer to the surface layer in the case of lower pressure compacts; and closer to the center line of the thickness in the case of higher pressure compacts. So, it can be believed that grain growth requires the critical deformation of the powder particles, but not of the mass of the specimen as a whole. Unfortunately, we can not control and measure the critical deformation of individual powder particles accurately enough to find out the most favorable condition for individual grain growth.

In the case of higher pressure compacts, the powder particles of surface layer received more deformation than the critical amount; and in the case of the lower pressure compacts, the powder particles of the surface layer received less than the critical amount, so the critical deformation always was located at different positions between the center to the surface layer for different pressures. And also this critical deformation range or layer has different width for different pressures.

From the data of this research, the grain growth curve (Fig. 16) always showed the larger grain sise in the pressure range of 8 to 12 tsi not only after sintering, but also after repeated deformation and annealing. So we can say, the chief factor of the grain growth of powder compacts is the compressing pressure.

<u>Speed of compressing</u> ——— The speeds of punch advance were limited by the pressures used on the machine. In this research, the different speeds of pressing was tried on several specimens with same pressure. The rate of speed (0.3 in./min.) produced 'blowholes' in the specimens after sintering; these 'blowholes' not only make the surface

of compacts uneven, but also are very harmful for grain growth. Because the faster compressing speed will promote the larger bridge formation, and the air entrapped cannot be evolved easily through the higher density surface layer during sintering, the surface of compacts showed a dome like form , with some caves under it. During cold-working and annealing, these caves cannot be compacted together, due to the cause that the inner surface of the caves may be deeply oxidized. After annealing, the cross section showed many crack lines. These lines would limit the grain growth. So we can conclude that the lower the pressing speed, the easier will be the grain growth, if other conditions are the same.

Volume change and density change ——— In this research, the volume change of powder compacts was studied, as shown in Fig. 14. After compressing, the volume of compacts decreases with increasing pressure, but after sintering, the relation is just the reverse, the lower the compressing pressure, the smaller the volume. In other words, after sintering, the amount of volume shrinkage was decreasing with increasing pressure from the range of 2.45 to 20 tsi; from 20 to 25 tsi the volume had not much change; above 25 tsi, the volume of compacts showed expansion instead of shrinkage. The lower the pressure is, the more the air will be entrapped after compressing. During sintering, the air was evolved, thus the volume was reduced.

As shown in the curve of Fig. 15, the tendency of density change is almost just the reverse as that of volume change. After pressing, the green density increased with increasing pressure; but after sintering, the lower the pressure was, the higher the density. It was directly due to the volume shrinkage. As explained by Paul Schwarzkopf(32), the powder pressed at extremely high pressure frequently shows a decrease of density with increasing sintering temperature. In these cases, the density produced by compression is very high and is reduced during sintering by growth due to the escape of gases. Schwarzkopf gave three theoretical curves for different pressures; these curves show the high pressure compacts decrease density with sintering temperature, the density of low pressure loose compacts increase with sintering temperature, the moderate pressure compacts density increased too.

B. Deformation and grain size

The grain size observed in the copper wire of Experiment 2, after the wire was annealed at 950°C for five hours, is about 10 times larger than that observed in the tensile deformation for both cases are 2% elongation. It indicates that the deformation may be not large enough in the case of tensile bars. Since the difference between the powder compacts and cast metal is the porosity, so for grain growth, the percent of deformation of powder compacts

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(32) Schwarzkopf, P., Powder Metallurgy, New York, Macmillan Co. 1947, p. 103

may be a little more than that for cast metal, since the greater the porosity, the greater the amount of deformation required to deform the grain itself into a definite amount. In the case of the powder, compact, when deformation takes place, it is very possible that some of the deformation is just enough to reduce the porosity, the crystalline mass thus not receiving the true percent of deformation required for subsequent growth.

The grain size observed in the flat samples of Experiment 1, by the method of repeated deformation and annealing is much larger than that observed in the tensile bars of Experiment 3 by the method of single deformation and annealing. It shows that the grain will be grown larger and larger when repeated mechanical and thermal energies are applied. In each cycle, the mechanical energy will create a potential difference between grains, thus a tendency for rearrangement is established and the thermal energy will tend to increase the vibrational energy so as to bring about this arrangement.

C. Limitation of grain growth

It is obvious that the thickness of specimens is the limit of grain growth in the case of cast metals, and in the case of powder compacts, the grain growth is limited not only by the thickness of the specimen but also by the porous crack lines present in the compacts which are prepared under a very low pressure and a higher speed of compressing.

The other conditions of limitation of grain growth for both the cast metals and powder compacts is the equilibrium factor. That means if the specimens show uniform grain size over the entire area of the sample, then grain growth will be stopped, even with further increase of annealing temperature and time. Because during the condition of equilibrium, the surface tension of each grain is the same, the energy content is the same, therefore, there will be no crystal which could absorb or be absorbed by any other crystal.

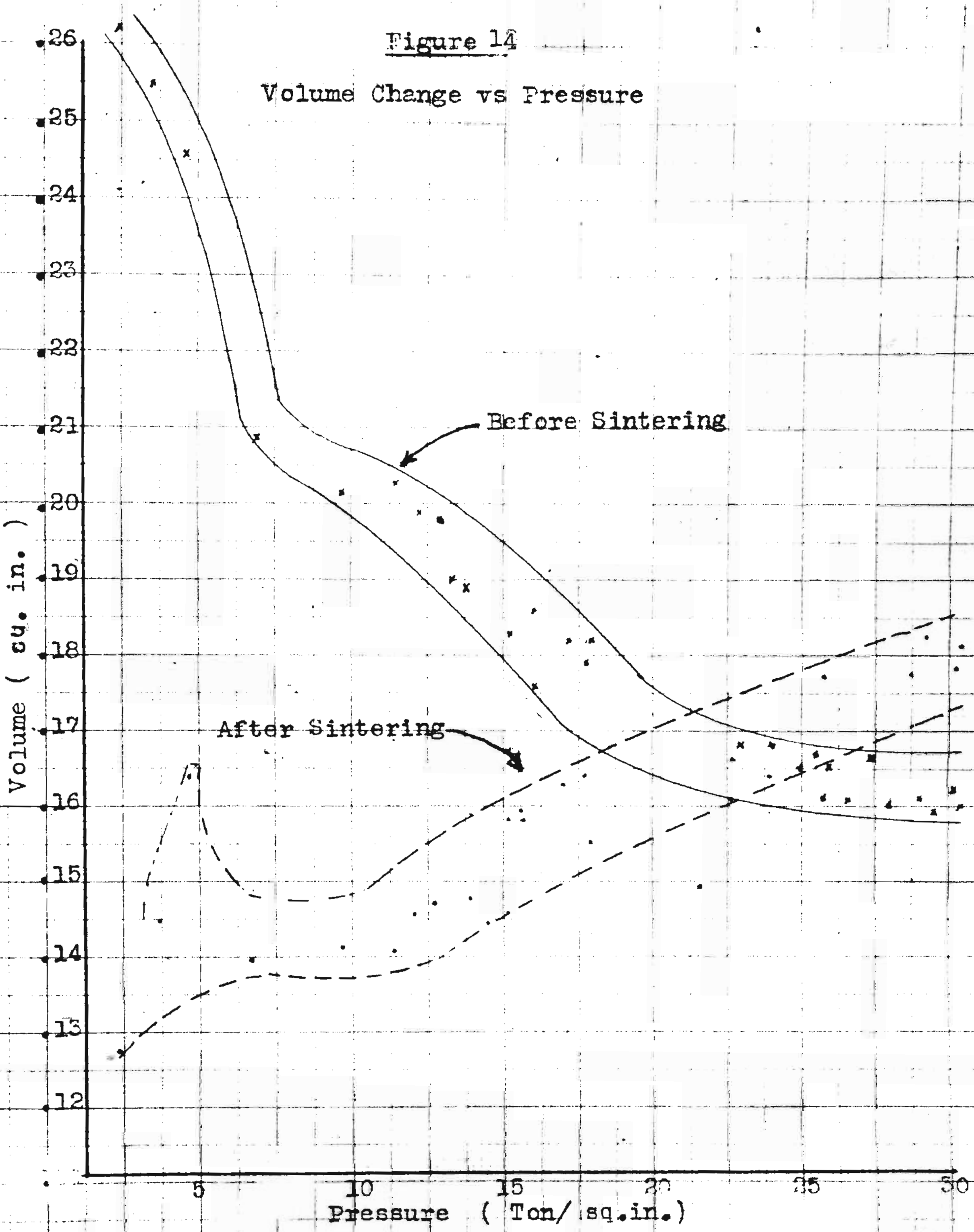


Figure 14

Volume Change vs Pressure

Figure 15

Density Change vs Pressure

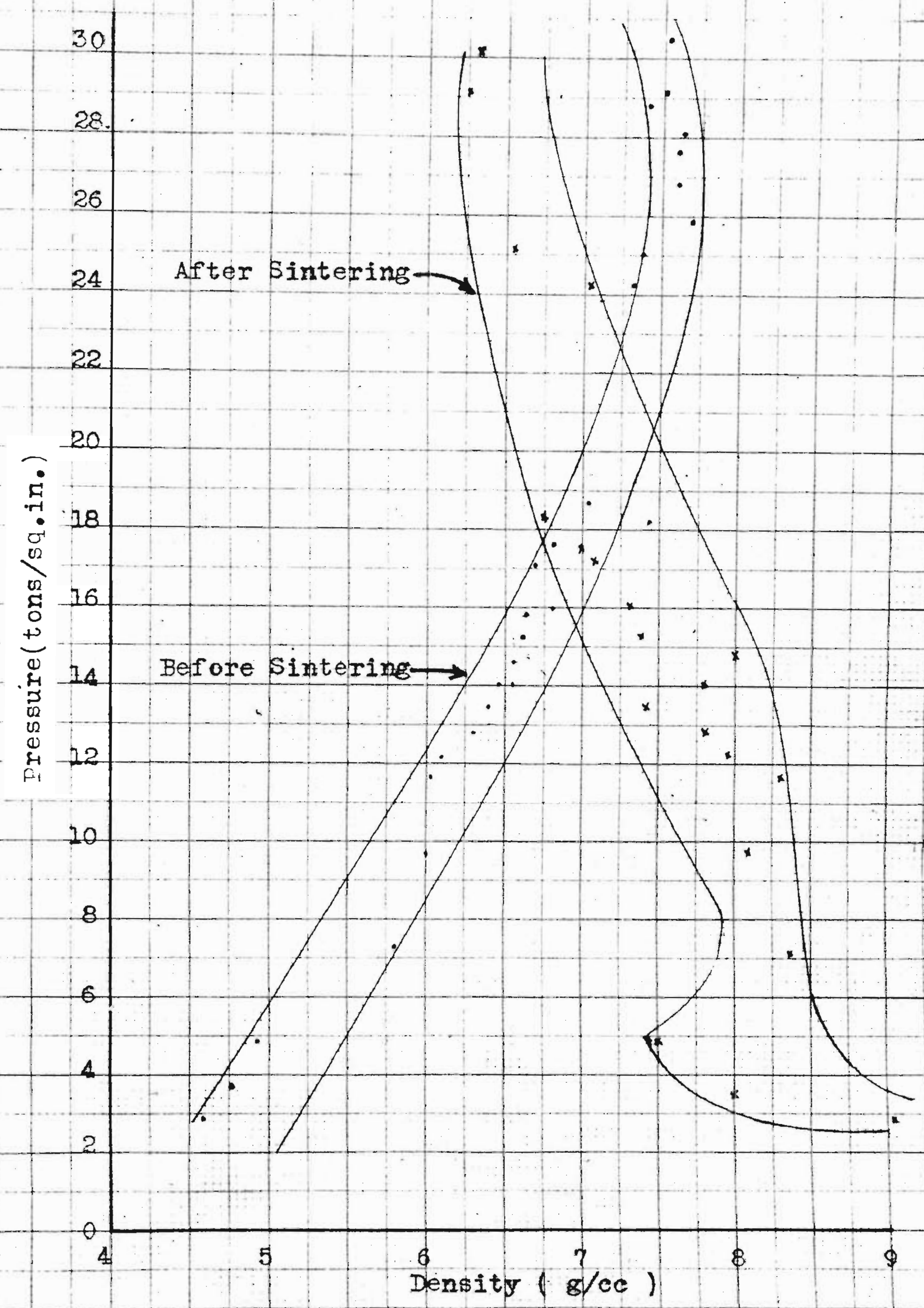
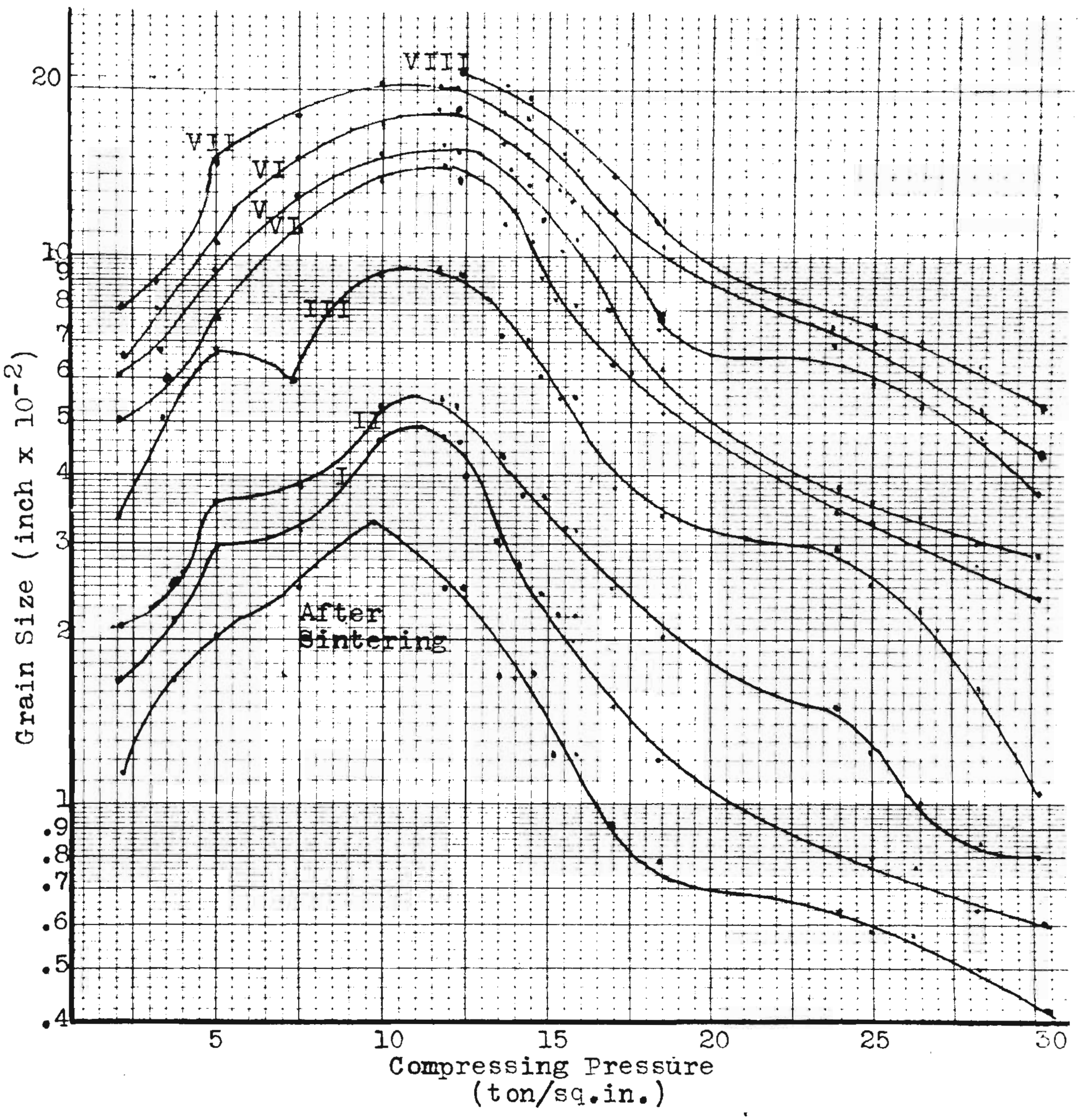


Figure 16

Grain size with pressure



I, II, III, IV, V, VI, VII, and VIII---are number of annealing

VIII

Conclusion

From the results obtained by this investigation, it appears that the favorable conditions for the grain growth of copper powder of the type and composition used are as follows:

(1) A compressing pressure of 8 to 12 tsi.

(2) A sintering temperature of 950°C.

(3) An annealing temperature of 950°C.

(4) The longer the annealing time, the better.

(5) The longer the sintering time the better.

(6) Slow speeds of pressing are preferred to faster ones.

Apparently the copper powder metal compacts need more cold working to give the same size recrystallized grains than cast copper.

Cyclic cold working and annealing tends to give larger recrystallized grains than any of the other methods tried.

Pores after sintering were almost uniformly distributed at the boundaries of the grains. After cold working and annealing, they tended to join one another to form larger pores or were included within the new formed grains. Extremely large pores gave rise to crack lines on the specimen and this limited grain growth.

No single crystals over the entire gauge length of the tensile sample were formed, but techniques were developed that tended to show that under the proper conditions of pressing and sintering, this might be accomplished.

IX

Summary

From the experiment of this research, to prepare the single crystals from copper powder by powder metallurgy method followed by compression, sintering, deformation, either elongation or compression and annealing appears to be feasible. It may be true that the powder compacts need more relative plastic deformation to cause grain growth than the cast metals.

The favorable temperature for the grain growth of copper powder compacts during sintering or annealing is 950°C (1710°F) and the favorable compressing pressure for the grain growth of copper powder compacts fall within range of 8 to 12 tsi.

The process of recrystallization and grain growth in the copper powder compacts and the copper wire appears similar; either the large crystals absorb the smaller crystals or the smaller crystals absorb the larger crystals, and both showed characteristic twin crystals.

The porosity of powder compacts is not harmful to grain growth, because the pores can be included in new grains formed from recrystallization and grain growth. Very large pores, however, are undesirable.

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