2-18-1944

Hardness and Electrical Resistivity of Copper-Iron Powder Metal Compacts

Fred A. Foyle Jr.

Recommended Citation

Foyle, Fred A. Jr., "Hardness and Electrical Resistivity of Copper-Iron Powder Metal Compacts" (1944). *Bachelors Theses and Reports, 1928 - 1970.* Paper 196.

HARDNESS AND ELECTRICAL RESISTIVITY OF COPPER-IRON POWDER METAL COMPACTS

by

Fred A. Foyle, Jr.

A Thesis
Submitted to the Department of Metallurgy
in Partial Fulfillment of the
Requirements for the Degree of
Bachelor of Science in the Metallurgical Engineering

MONTANA SCHOOL OF MINES
Butte, Montana
February 18, 1944

HARDNESS AND ELECTRICAL RESISTIVITY OF COPPER-IRON POWDER METAL COMPACTS

by

Fred A. Foyle, Jr.

A Thesis
Submitted to the Department of Metallurgy
in Partial Fulfillment of the
Requirements for the Degree of
Bachelor of Science in Metallurgical Engineering

MONTANA SCHOOL OF MINES
Butte, Montana
February 18, 1944

TABLE OF CONTENTS

Page

INTRODUCTION 1

CHOICE OF METAL POWDERS 3

THEORY AND APPARATUS 5
- Strength of Compresses
- Mixing the Powders
- The Press
- The Sintering Furnace
- Rockwell Hardness Tester
- Electrical Resistance Measurements

PROCEDURE 15

DATA AND GRAPHS 18

EXPERIMENTAL RESULTS 29

CONCLUSION 33

SUGGESTIONS FOR FURTHER WORK 35

BIBLIOGRAPHY 36

ACKNOWLEDGEMENTS 38

INTRODUCTION

Powder metallurgy may be defined as "the art of making, forming, and treating metal powders and their products; the products are essentially made without melting"*[4], although this definition includes the following four fields, only the last three will be discussed in this paper: (1) preparation of the powders, (2) compressing the powder into the desired shape, (3) heat treating, and (4) study of the properties of the finished product.

Although powder metallurgical methods have been used for years to fabricate tungsten and platinum, very little scientific data have been recorded until the beginning of this century. A large percentage of all commercial production at present is based upon past practice rather than upon scientific knowledge. Only recently have great advances been made in scientific knowledge through laboratory research. New products are being produced daily by these methods, because it not only saves precision machines, but precious man-hours as well. The high production costs of the metallic powders and the limitations in product size are the two great disadvantages which must be overcome before powder metallurgy can be considered

*Numbers refer to references in the bibliography.

a great competitor of casting and other older fabricating methods.

The entire first semester of the author's thesis work was devoted to library research, to acquaint the author with the terms used, the theory, and much of the previous research in the field of powder metallurgy.

The original plan of the experimental work was to compare the electrical resistance of copper-iron powder compacts with those of cast alloys of the same composition, but due to the short time available for actual laboratory work and the many experimental difficulties encountered, it was impossible to complete this procedure. The copper-iron compacts were made and their hardness and electrical resistances were measured but no work was attempted with the cast alloys.

CHOICE OF METAL POWDERS

Copper and iron were chosen as the two metals to be investigated. Copper is the best conductor of electricity among the cheap, plentiful metals, and if it were possible to produce a copper-iron compact that would retain the high electrical conductivity of copper and still have the hardness and wear-resistance of iron, it would form a very desirable alloy for such electrical applications as contact points, which are subject to great wear. These two metals, which are very difficult to alloy by melting, can easily be mixed in the powder form and treated to give a homogeneous alloy of any composition.

Of prime importance in this process is the size and surface condition of the powders. Even a very light film of oxide on the surface of each particle may greatly reduce the strength of the compress and may have other very harmful effects. The copper powder used was of commercial grade, and had been sealed in a container to prevent oxidization. The iron powder was of the same grade, but probably contained considerably more oxides as its container had previously been opened. The copper powder was much the finer, being about 90 per cent minus 300-mesh, while the iron powders were only about 50 per cent minus 300-mesh. Both powders were 100 per cent minus 100-mesh.

A first set of alloys was made with highly oxidized copper powders. These were found to be distinctly inferior to the compresses made later with unoxidized powders. This first set, although pressed at the same pressure and conditions, used later, crumbled very easily, and had very high electrical resistance. Therefore, this entire series was disregarded, in preference to the set later made with fresh unoxidized copper powders. The same iron powder was used throughout.

THEORY AND APPARATUS

Strength of Compress

The strength of a powder compact is proportional to the forces which hold the individual particles together. These particles are held together by mechanical inter-locking and cohesion.

Mechanical inter-locking depends upon the irregularities and size of the individual particles making up the compress and upon the pressing pressure. The smaller and more irregular the particles and the greater the pressure, the stronger and harder the resulting compact will be.

Cohesion is the principal force holding the particles of a compress together. Although very little scientific data is available on the subject, the cohesive force depends both upon the area of contact of one particle with another, and upon the properties of the metals concerned. The greater the pressure used in pressing, the greater will be the area of surface contact between particles, and the stronger the resulting compact. Sintering also increases the cohesive force between particles, but no satisfactory explanation is given for this phenomenon. Numerous theories have been offered in explanation of cohesion but, up to the present time, scientific research has not progressed far enough to formulate any definite conclusions. The reader is referred to other more extensive works[4,5] for further information on this subject.

Mixing the Powders

After the correct amounts of the two metal powders were carefully weighed to give the desired percentage compositions in the alloys, they were placed together in test tubes and sealed with rubber stoppers. The test tubes were then placed on a small set of rolls (Page 26, Plate 2) and allowed to rotate for approximately four hours, to insure a thoroughly uniform mixture for pressing. This mixture was then ready to be pressed into the desired shape.

The Press

The press, a picture of which may be found on (Page 26, Plate 1) was made in the shops of the School of Mines and is fairly successful in its operation. It consists of a base plate to which are welded four uprights, a hydraulic jack with a gauge on which the total pressure applied to the piston is registered, three horizontal plates attached to the uprights (two of which hold the cylinder while the third one guides the piston), a piston, a cylinder, and a cap for the cylinder. The steel cylinder, or mold, which is four inches long and two and one-half inches in diameter, has a half inch hole drilled lengthwise through its center. Into this hole passes the upper end of the pressing piston. The mold has two ports through which gas may be passed to control the pressing atmosphere, and is surrounded by an electric heating coil for hot pressing. Neither the ports nor the coil were used, since all compresses were made in an air atmosphere and at room temperature.

The powder, after being completely mixed was poured into the cylinder, and the cap put in place, covering the top end of the cylinder. The piston was then forced up by use of the jack until the desired pressure was obtained. This pressure was maintained for three minutes after which the compact was ejected from the mold.

This operation, simple as it may appear, caused considerable trouble. After making and ejecting a compress it was found difficult, and sometimes impossible, to remove the piston from the cylinder. Graphite and oil were used as lubricants, but difficulties were still encountered. The cylinder, which had become nicked and scarred on the interior, caused the piston to stick. It was finally necessary to make two new pistons to complete this work. Before any further work is attempted with this apparatus, both a new cylinder and piston should be obtained.

The Sintering Furnace

The sintering of all the compacts was carried on in an electrical resistance-type furnace (Page27, Plate 3) at 700°C. An attempt was made to keep the compresses from oxidizing at this temperature by covering them with charcoal. This proved to be futile, as after sixteen hours at 700°C, all of the carbon had been oxidized, as had the outer surfaces of the compacts. A previously calibrated thermocouple was used to measure the sintering temperature.

Rockwell Hardness Tester

The hardness of each compress was measured using a one-sixteenth inch steel ball point and a fifteen kilogram load on the Rockwell Superficial Hardness Tester. With this combination it was possible to record the hardness of all the compresses within the limits of one set of gauge readings. Three hardness readings were taken on both the top and bottom of all compresses. In this way two average hardnessess, which are recorded and graphed, were obtained for each compact.

Electrical Resistance Apparatus

The electrical resistance apparatus consisted of a galvanometer, two small known resistances of the order of one ohm each, two large variable resistances -- one 100 ohms and the other about 2000 ohms, a 20,300 ohms protective resistance for the galvanometer, a double-pole double-throw switch, a single throw switch, an Edison Storage Battery, two silver contact plates and a bench vise in which the plates and compacts were clamped. A wiring diagram of the apparatus appears on (Page14) and a photograph on (Page27, Plate 4).

The method used for measuring the resistance of the compacts was originated by Dr. Shue; the following description is taken from a thesis[1] written by John Fitzpatrick. It was necessary to make a few minor changes before this apparatus was satisfactory for the present work, since a galvanometer of higher sensitivity than that in Fitzpatrick's work, was used.

"The principle upon which the setup worked is based upon comparing the I R drop across the compact in terms of a deflection on the galvanometer with the

I R drop across one of the small known resistances, which is measured by a second deflection on the galvanometer scale. (Referring to the wiring diagram Page 14.)

X is the resistance of the compact.

S is a small resistance and equals for all measurements, the value 1.2721 ohms.

T is a second small resistance, whose value is 1.2690 ohms.

R is the large, variable resistance of the order of 100 ohms.

I is the current in the main circuit."

(U is a variable resistance of the order of 2000 ohms used only to reduce the total current in main circuit).

"When the double pole switch is thrown to the left, across the resistance of the compact, there is registered on the galvanometer a deflection, D_1. When the switch is reversed, across the resistance, S, a deflection D_2 is registered. The relation is then as follows:

$$\left.\begin{array}{ll} & IX = KD_1 \\ \text{and} & I_sS = KD_2 \end{array}\right\} \text{------------}(1)$$

K is the deflection constant of the galvanometer.

I_s is the current flowing through S and R which are in series, and both of which are in a parallel with T.

From the above relationship the following equation may be written:

$$\frac{IX}{I_s S} = \frac{D_1}{D_2} \text{ -------------(2)"}$$

(Since, in a parallel circuit, the current divides inversely as the resistance, and the voltage drop across each branch must be equal, it follows:

Because $E_s = I_s (R + S); E_T = (I - I_s) T$; and $E_s = E_T$

Therefore $I_s (R + S) = (I - I_s) T$

$I_s (R + S + T) = I T)$

$$I_s = \frac{I\ T}{R + S + R} \text{ -----(3)}$$

"With this relation established between I and I_s, by substitution in equation (2),

$$\frac{IX}{\frac{T}{S + R + T} IS} = \frac{D_1}{D_2}$$

$$\text{And } X = \frac{D_1}{D_2} \frac{ST}{S + R + T} \text{ -----(4)}$$

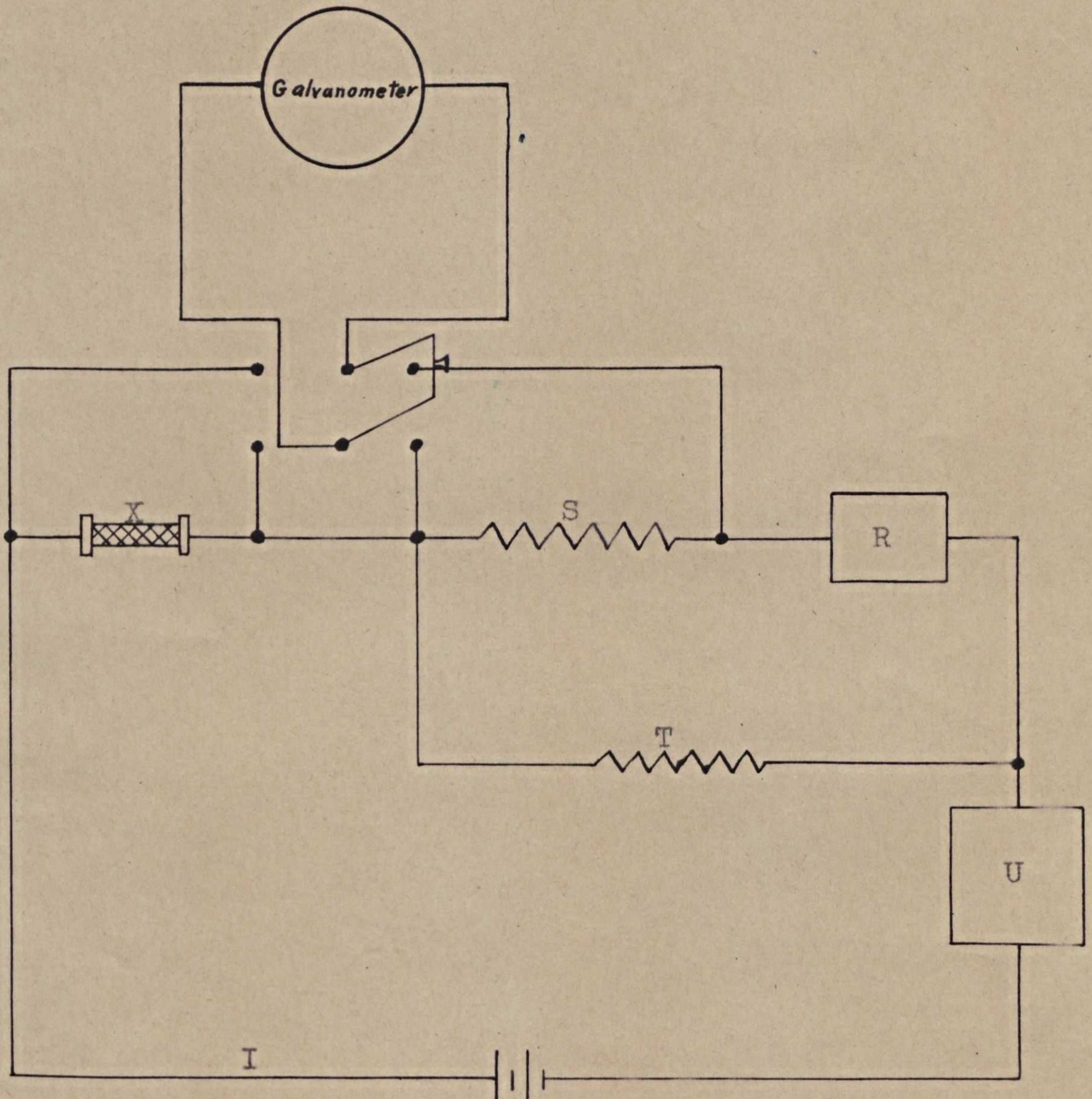
"Equation (4) was the one used to determine the resistance X of the compact. S and T were fixed, D_1 and D_2 were measured, and R was fixed, making the calculation possible.

"When the resistance of a compact was measured, the compact was placed between the silver plates on the jaws of the vise and the jaws tightened the ends of the compact. The switch was then thrown and D was read on the galvanometer. This was recorded and the switch was thrown to the opposite set of poles and D_2 was read. To check this reading, the compact was removed from the vise and turned end for end and replaced. Readings were again taken in the same manner."

This same procedure was followed, throughout the laboratory investigation, although it was necessary to use only 2.8 volts and a large 2000 ohm variable resistance between the current source and the apparatus to further cut down the voltage. Otherwise, since a very sensitive galvanometer was used, the deflections were too great to be conveniently and correctly read.

The compacts, as they came from the mold, did not have perfectly flat smooth ends, and therefore, it was necessary to grind the ends of each compact down to the smoothness of 00 emery paper, to insure good electrical contacts with the silver contact plates.

These plates, after a backing of sponge rubber was cemented to them, were fastened to the jaws of a bench vise. The sponge rubber served two purposes. First, it was a good electrical insulator, and, second, it allowed the plates to form a more nearly perfect contact with the end of the compact. After sintering, due to oxidization of the surface of the compacts, it was found necessary to repolish these ends to insure good contact.



ELECTRICAL RESISTANCE MEASURING APPARATUS

PROCEDURE

A series of fourteen compacts were made with compositions varying from 100 per cent copper to 100 per cent iron. All of the compresses were pressed at 40 tons per square inch, no attempt being made to control the pressing atmosphere, and were held at this pressure for three minutes. The composition of the compresses was as follows:

No.	Compositions	
1	100% Cu.	0% Fe.
2	95% Cu.	5% Fe.
3	90% Cu.	10% Fe.
4	85% Cu.	15% Fe.
5	75% Cu.	25% Fe.
6	65% Cu.	35% Fe.
7	55% Cu.	45% Fe.
8	45% Cu.	55% Fe.
9	35% Cu.	65% Fe.
10	25% Cu.	75% Fe.
11	15% Cu.	85% Fe.
12	10% Cu.	90% Fe.
13	5% Cu.	95% Fe.
14	0% Cu.	100% Fe.

After the compresses were made, the hardness and electrical resistance of each was measured and recorded. Before the electrical resistance was measured, it was necessary to polish both ends of each compact with 00 emery paper to insure a smooth surface that would produce a good electrical contact. It was also necessary to measure both diameter and length of each compress so that the resistivity could be calculated.

These samples were then sintered at 700°C for sixteen hours in a clay crucible. The samples were completely covered with charcoal to prevent excessive oxidization, but in spite of this precaution, the compresses became highly oxidized, especially near the surface. Although this oxidized zone was very thin, it was necessary to polish the ends of these samples before either hardness or electrical resistance could again be measured.

Two compresses--both 45 per cent iron, 55 per cent copper--were polished and micro-photographed. The first specimen had not been sintered, while the second one was sintered for sixteen hours at 700°C.

The unsintered specimen (Page 28, Plate 5,) has considerably more large voids than has the sintered compact (Page 28, Plate 6). This would explain both the greater hardness and the decreased resistivity observed for the sintered compact. No actual alloying of the copper and iron was observed at the contact of the individual particles of these metals even after sintering--although a limited zone of alloying may have existed. It was quite difficult in the micro-photographs of the compacts to distinguish between the copper and iron particles present. When examining these sections under the microscope these difficulties were eliminated due to the differences in color of the two particles, although this does not show up well in the black and white photographs.

DATA

UNSINTERED COMPACTS

Composition	Length[1] (cm)	Area[2] (cm)^2	X Ohms	Resistivity Ohm/cm cube
Cu. 100% Fe. 0%	1.55	1.30	0.0016	0.00134
Cu. 95% Fe. 5%	1.50	1.30	.0027	.00231
Cu. 90% Fe. 10%	1.517	1.30	.0021	.0018
Cu. 85% Fe. 15%	1.526	1.30	.0018	.00153
Cu. 75% Fe. 25%	1.545	1.30	.0025	.0021
Cu. 65% Fe. 35%	1.647	1.30	.0054	.00425
Cu. 55% Fe. 45%	1.712	1.30	.0176	.0133
Cu. 45% Fe. 55%	1.797	1.30	.0634	.0459
Cu. 35% Fe. 65%	1.790	1.30	.1861	.1563
Cu. 25% Fe. 75%	1.842	1.30	.180	.1502
Cu. 15% Fe. 85%	1.766	1.30	.199	.1692
Cu. 10% Fe. 90%	1.996	1.30	.371	.3412
Cu. 5% Fe. 95%	1.785	1.30	.401	.3712
Cu. 0% Fe. 100%	1.827	1.30	.199	.1692

1. Length of the actual compresses increased uniformly with the amount of Fe. present but it was necessary to polish some compacts more than others. This accounts for the non-uniformity in the lengths recorded above.

2. Area is the total cross-sectional area of the compact.

COMPRESSES SINTERED FOR SIXTEEN HOURS AT 700°C.

Composition		Length (cm)	Area (cm)^2	X Ohms	Resistivity Ohms/cm cube
100% Cu.	0% Fe.	1.535	1.34	0.0001	.000088
95% Cu.	5% Fe.	1.495	1.34	.0001	.000090
90% Cu.	10% Fe.	1.523	1.34	.0001	.000088
85% Cu.	15% Fe.	1.585	1.34	.00016	.000135
75% Cu.	25% Fe.	1.524	1.34	.0001	.000088
65% Cu.	35% Fe.	1.651	1.34	.00014	.000113
55% Cu.	45% Fe.	1.711	1.34	.00016	.000125
45% Cu.	55% Fe.	1.800	1.34	.0002	.000149
35% Cu.	65% Fe.	1.802	1.34	.0009	.00067
25% Cu.	75% Fe.	1.837	1.34	.0009	.00066
15% Cu.	85% Fe.	1.741	1.34	.0009	.00069
10% Cu.	90% Fe.	1.865	1.34	.0018	.00130
5% Cu.	95% Fe.	1.753	1.34	.0026	.00199
0% Cu.	100% Fe.	1.753	1.34	.0026	.00194

ROCKWELL HARDNESS TESTS[1] UNSINTERED COMPACTS

Compositions		Top	Bottom	Average[2]
100% Cu.	0% Fe.	62.7	71.7	67.2
95% Cu.	5% Fe.	51.7	64.7	58.2
90% Cu.	10% Fe.	49.7	64	56.7
85% Cu.	15% Fe.	52.3	64	58.1
75% Cu.	25% Fe.	50	65.6	57.8
65% Cu.	35% Fe.	49	66	57.5
55% Cu.	45% Fe.	44	65	54.5
45% Cu.	55% Fe.	35.4	60	47.7
35% Cu.	65% Fe.	40.7	62.7	51.7
25% Cu.	75% Fe.	29	60	44.5
15% Cu.	85% Fe.	19.5	46.3	32.9
10% Cu.	90% Fe.	26.3	40.3	33.3
5% Cu.	95% Fe.	32	48.7	40.3
0% Cu.	100% Fe.	40	51.7	45.8

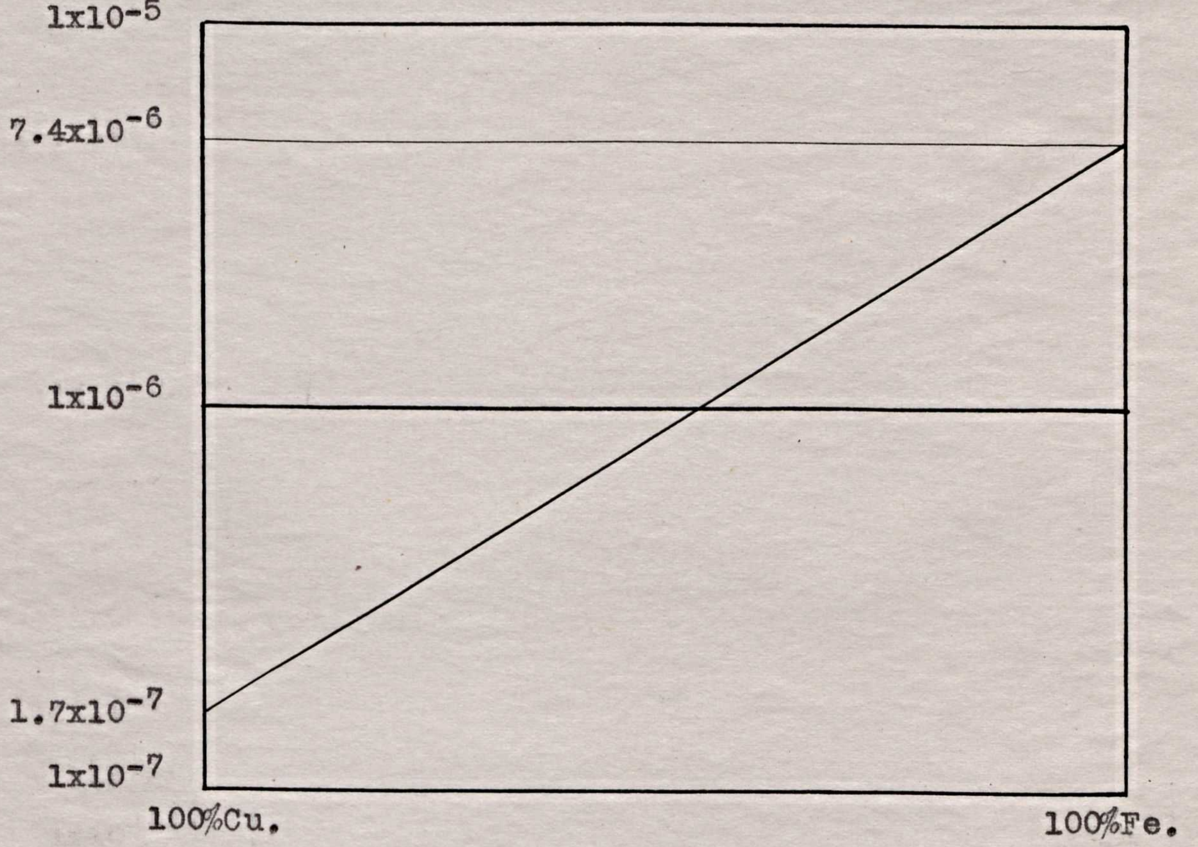
1. A Rockwell Superficial Hardness Tester with a 15 Kilogram load and 1/32 round steel ball was used to make all hardness readings.

2. Three hardnesses were taken on the top of each speciman and three more on the bottom. These were then averaged to get the top and bottom averages, and these in turn were averaged again to give the overall average for each compact.

ROCKWELL HARDNESS TESTS

(COMPACTS SINTERED FOR 16 HOURS AT 700°C)

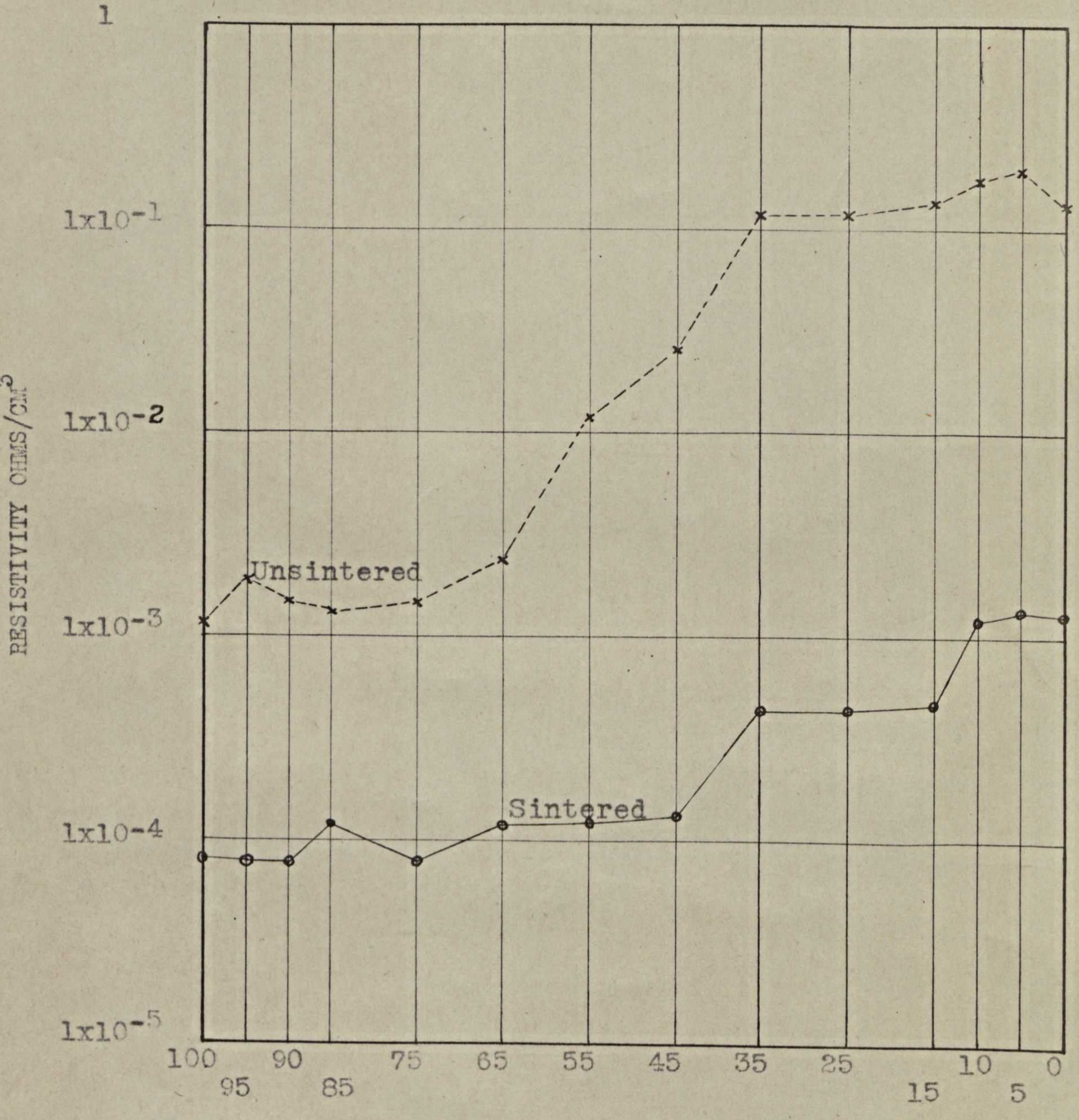
Composition	Top	Bottom	Average
100% Cu. 0% Fe.	30	37.2	33.6
95% Cu. 5% Fe.	69.6	63.6	67.6
90% Cu. 10% Fe.	66	68.6	67.3
85% Cu. 15% Fe.	68	69.	68.5
75% Cu. 25% Fe.	69.3	70.6	69.9
65% Cu. 35% Fe.	70.3	67.6	68.9
55% Cu. 45% Fe.	66	69.6	67.8
45% Cu. 55% Fe.	69	72.6	70.8
35% Cu. 65% Fe.	72	75	73.5
25% Cu. 75% Fe.	76	75	75.5
15% Cu. 85% Fe.	75.7	74.3	75
10% Cu. 90% Fe.	71.3	75	73.1
5% Cu. 95% Fe.	76	76	76
0% Cu. 100% Fe.	80	81.3	80.7



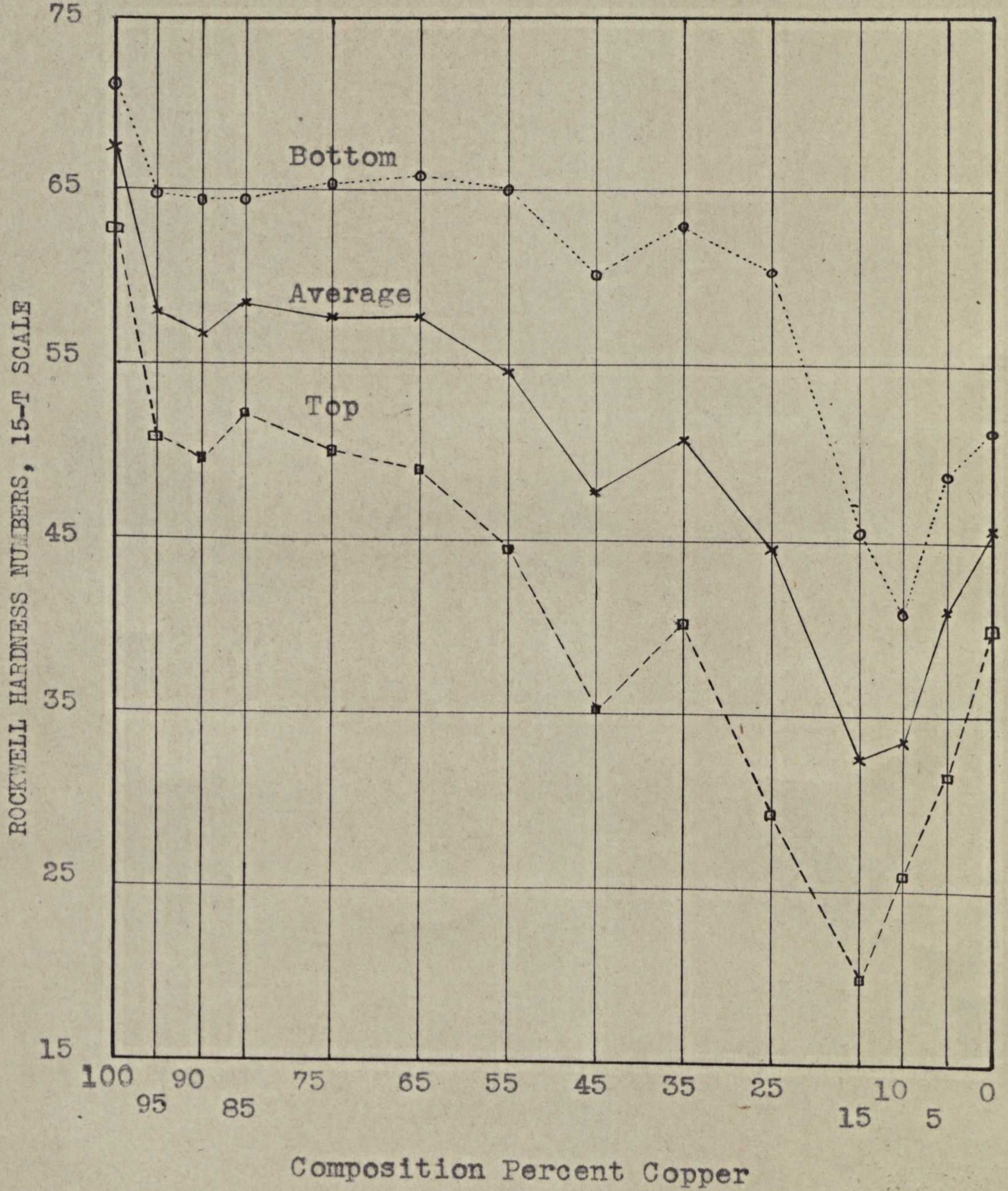
THEORETICAL RESISTIVITY*

---

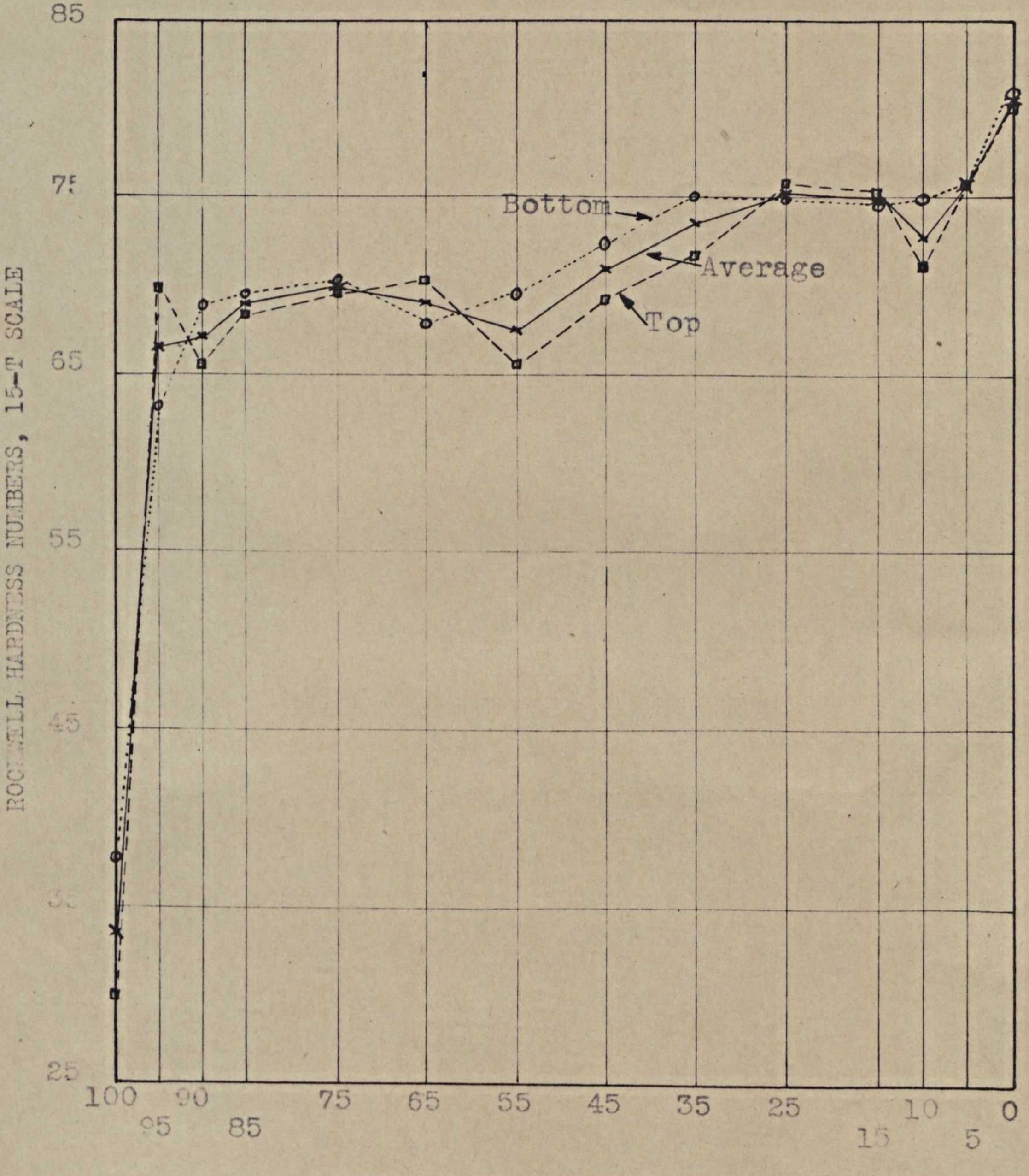
* We may draw this theoretical graph because the properties are averaged due to the formation of a purely mechanical mixture rather than a solid solution.



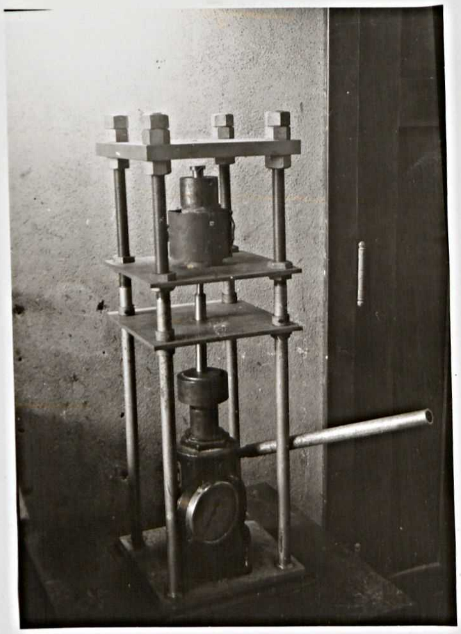
MEASURED RESISTIVITY OF COMPACTS



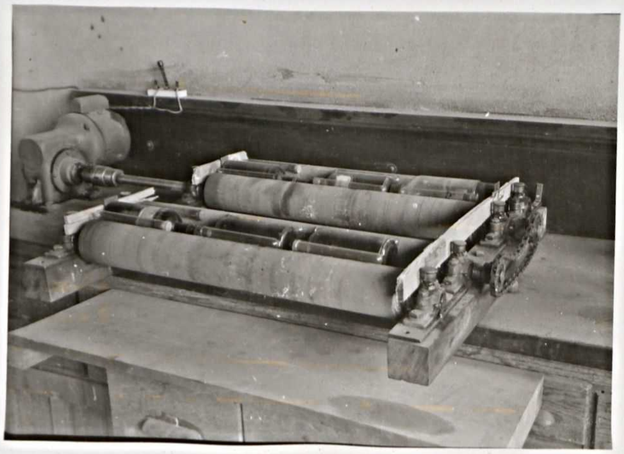
MEASURED HARDNESS BEFORE SINTERING



MEASURED HARDNESS AFTER SINTERING



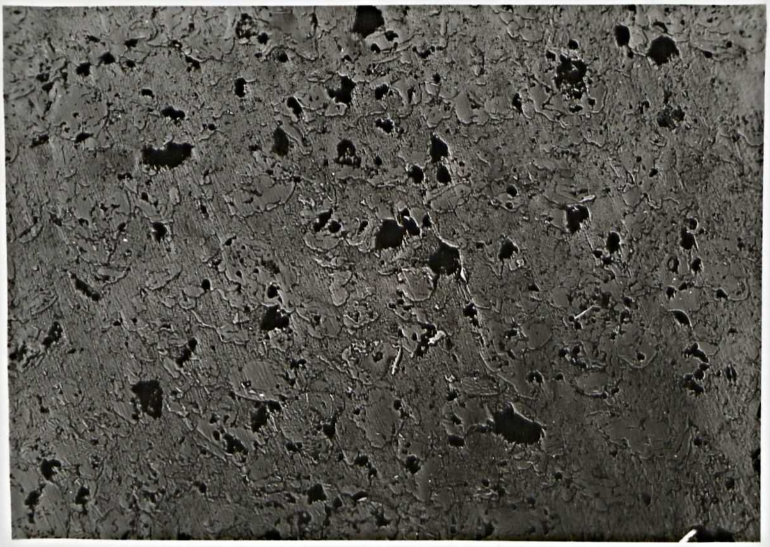
The Powder Press
Plate 1



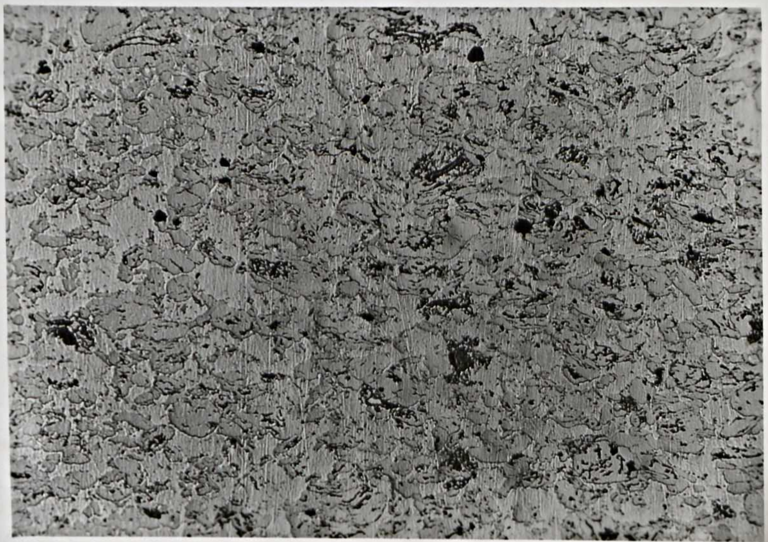
The Rolls for Mixing the Powders
Plate 2

The Sintering Furnace
with Thermocouple
Plate 3

The Electrical Resistivity
Measuring Apparatus
Plate 4



45% Iron, 55% Copper
Unsintered Compact
100 X, Unetched
Plate 5



45% Iron, 55% Copper
Sintered Compact
100 X, Unetched
Plate 6

EXPERIMENTAL RESULTS

Various pressures were used on a series of preliminary compacts to determine a workable pressure. After examining these samples, it was found that forty tons per square inch gave the most satisfactory results. With this pressure, the 15 gram compresses varied in length from 1.55 to 2 centimeters. The pure copper, being soft, produced the shortest compacts, and as the amount of iron increased, the length of the compress also increased, provided the same pressure was applied. This was to be expected, since the iron particles were larger in size and were much less plastic than the copper particles. Since the specific gravity of the iron is less than that of copper, this would also tend to make the high iron compress longer than the high copper compresses since a given weight of iron would occupy more volume than the same weight of copper and the total weight of the compacts was constant. After sintering, both the diameter and length of compact increased, although no satisfactory explanation for this expansion can be given.

The hardness of the compacts before sintering, as measured on a Rockwell Superficial Hardness Tester, was found to vary about fifteen points between top and bottom of each compress. In every case, the bottom, where the pressure was applied, was considerably harder than the top. This was to be expected since the pressure throughout a powder is not equal, but is the greatest at the point of application of the force. This difference in hardness could be overcome to some extent by having more than one point at which the pressure is applied.

The high copper compresses were found to be much harder before sintering than were the high iron ones. This was probably due to the ability of the copper to deform under pressure to form a compact, work-hardened matrix in which the iron particles were imbedded. The iron particles were not as easily deformed, and therefore, the high-iron compacts consisted of a looser, less coherent mass, with insufficient copper for good bonding, resulting in a softer compact.

The 100 per cent copper compact was softer after sintering than before, due to the annealing process, which removed the effects of previous strain-hardening. The temperature and time of anneal were probably sufficient to give this metal a full anneal. All of the other compacts increased greatly in hardness after sintering, and the more iron present, the harder each compress became. Before sintering the high-iron alloys had large voids, but after sintering these voids appeared to be more numerous but much smaller in size. The sintered alloys, therefore, had more of the powder particles in contact with each other, and therefore, cohesive forces were greater, so that a much stronger, harder material resulted. After the cohesive forces were great enough to hold the particles together, the greater inherent hardness of the iron particles accounted for the increased hardness of the high iron compacts over the low iron compacts.

The resistivity of the compacts before and after sintering approximately followed the theoretical curve, but the resistances in each case were much greater than these theoretical values. In the case of the 100 per cent copper compact, the resistivity was about 500 times the theoretical value for cast pure copper and that of pure iron was about 100 times the theoretical value for cast pure iron. The resistivity of the pure copper compact was found to be very close to those given in the literature[5] for pressed copper powders, but it was not possible to find any data on on the resistivity of pure iron compacts or of copper-iron compact.

The resistivity for both the sintered and the unsintered series was found to increase greatly with iron content above about 50 per cent. This was probably caused by poor electrical contact between individual particles, in the compress, due to the low plasticity of the iron particles as well as to the poor conductivity of the iron itself. The unsintered compacts, due to the larger proportion of voids, had a very much higher resistance throughout the series than did the sintered series.

CONCLUSION

The series of compresses made with highly oxidized copper powders was found to be very much inferior to the series made with unoxidized copper powder, showing that the surface conditions of the powders have a great effect on the properties of the finished compresses.

The 95 per cent copper, 5 per cent iron alloys were much harder than the 100 per cent copper alloy after sintering; yet the resistivity increased only slightly. With proper heat treatment, such as improved control of sintering time and temperature, this alloy might be hardened considerably more to form a very hard metal with high conductivity. This would have many uses in electrical apparatus that is subject to wear.

The hardness and resistivity of the compacts apparently do not have any direct relationship. In the unsintered compacts the hardness decreased with increasing percentages of iron while the resistivity increased. The sintered compacts showed an increase in both hardness and resistivity with increasing iron content.

Sintering for sixteen hours decreased the resistivity of the high copper compacts ten fold and decreased the resistivity of the high iron compacts over one hundred fold. The low iron compacts, after sintering, had a very low resistivity.

SUGGESTIONS FOR FURTHER WORK

Although sufficient time was not available to carry out my original plan of comparing the hardness and electrical resistance of cast alloys with powder metallurgical compacts, the results that were obtained were consistent and promising. This comparison could very easily be made provided further time were available, and should show some very interesting results. Although I did not accomplish my goal, a good foundation was made upon which later workers may easily expand.

There are many other interesting problems in powder metallurgy. This is a new field and great opportunities are offered for research as many phenomena are still unexplained.

BIBLIOGRAPHY

1. Fitzpatrick, John M.
   "Electrical Resistivity Measurements of Powder Metal Compacts." Senior thesis, Montana School of Mines, May 1, 1942.

2. Raising, Robert
   "The Influence of Processing Variables on the Structure and Properties of Compresses and Heat-treating Metal Powders -- with Particular Emphasis on Copper-Nickel Alloys." Master's Thesis Montana School of Mines, May, 1940.

3. Shaw, Joseph E.
   "Production of Metal Compacts by the Powder Method with Special Reference to Copper- T in Alloys." Master's Thesis Montana School of Mines, May, 1940.

4. Wulff, John
   "Powder Metallurgy" Cleveland, The American Society for Metals, 1942.

5. Jones, W. D.

"Powder Metallurgy" London, England, Edward Arnold and Company, 1937

6. "Powder Metallurgy - Some important theoretical Aspects": Iron Age - Vol. 147, No. 16, April 7, 1941, Pages 23 - 27.

ACKNOWLEDGMENT

I would like to express my thanks to Professor Morton Smith and Dr. A. E. Koenig under whose guidance this work was carried on.

Fred A. Foyle Jr.
Fred A. Foyle, Jr.