INVESTIGATION OF SINTERED COPPER-MANGANESE-SULFUR POWDER COMPACTS

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SUMMARY

The following report presents the initial results of an investigation of the sintering and microstructural characteristics of copper rich, copper-manganese powder alloys containing small amounts of sulfur. Research on these materials has been undertaken to study the possibilities of their use as bearing materials, this application being based upon the preperties of MnS as exhibited in free machining steels, i. e. a material of low shear strength. Furthermore, the Cu-Mn system in the composition range investigated shows possibilities of age hardening; all of which leads to the possibility of a bearing material consisting of a high strength matrix with dispersed MnS or MnS-Cu_2S particles.

Initially, an attempt was made to introduce the sulfur as a sulfide of manganese on the basis that perhaps the copper would displace the manganese to a certain extent at the sintering temperature yielding a Cu-Mn solid solution and MnS-Cu_2S solid solution or mixture. The sulfide was prepared by heating a mixture of sulfur, manganese sulfate, and potassium carbonate. These compacts gave no indications of the desired structures but remained as MnS particles in Cu.

It was finally decided to use a pure Cu, Mn and S mixture with varying compositions of Cu and S. The first compacts were found to be very porous when examined after sintering.

As a remedy, the samples were presintered overnight at a temperature in the liquid sulfur range and then sintered at the desired high temperatures. A definite decrease in porosity was noted. The amount of sulfur was then reduced and the same procedure used. The resulting samples were practically solid and were much easier to polish.

The effect of time and temperature was determined on a mixture containing 58.5% Cu, 40% Mn, and 1.5% S. Definite diffusion was seen to take place; the amount of diffusion increased with both time and temperature.

Some samples were quenched from the Cu-Mn solid solution temperature range to see if a solid solution was formed. By comparison with similarly treated, slow cooled samples it was seen that a solid solution was definitely formed. This solid solution was believed to be a ternary solution dissolving both S and Mn in the Cu. It was also considered possible that a low melting eutectic might be formed consisting of some combination or combinations of all elements present as elements and/or sulfides.

Although this investigation did not completely cover any single phase of the system, it did, however, present some very interesting possibilities for further investigation.

INVESTIGATION OF SINTERED COPPER-MANGANESE-SULFUR POWDER COMPACTS

INTRODUCTION

The purpose of this investigation was to determine the possibility of using copper rich, copper-manganese powder alloys containing small amounts of sulfur for the formation of bearing materials. To be adaptable to this use the materials must exhibit free machining properties; that is, they must have a low shear strength. Low shear strength is necessary in bearings to prevent galling of metal in case the oil film, which is the actual bearing surface, breaks down. This low shear strength is one of the properties exhibited by MnS and for this reason sulfur was added to bring about formation of MnS. In the composition range investigated, the Cu-Mn system shows definite age hardenable characteristics; which leads to possibilities of a high strength matrix with embedded particles of MnS or MnS-Cu_2S.

A search for previous work on this system afforded only one reference which, due to the nature of the experiment, gave only limited information. This previous work by Guertler and Meissner[1] was with mixtures of Cu_2S and MnS that were melted and allowed to solidify. They found that two distinct layers formed on cooling; the upper being rich in MnS and the lower of pure Cu of mixed crystals of both Cu

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1. Meller, J. W.; A Comprehensive Treatise on Inorganic and Theoretical Chemistry; Vol. XII, page 397.

and Mn.

As such a large amount of segregation would cause very poor properties in a cast part; it was evident that only by using powdered materials could a suitable dispersion be obtained. By using powdered materials the equal distribution of all constituents could be obtained by thorough mixing of the powders before compacting; and no large amount of segregation would occur since the sintering temperatures would be well below the melting points of the metals present.

Both mixing technique and compacting pressure play an important part in the structural characteristics obtained from powdered metal parts. The shape and size of any parts made from powdered metals is limited by the peculiar characteristics obtained from of the lack of any lateral flow by the powdered metals when subjected to high pressures. For this reason no transverse cores may be placed in the part as can be done in poured castings; neither can a part with tapered sides be made from powdered metals and withstand any stress. Thus, the more uniform the shape, the better will be the properties.

The shape of the compacts used for this investigation was a thin disk about 1/8 inch thick and 0.2538 inch in diameter. To obtain these compacts the powdered mixture was placed in the die and subjected to a high pressure for 30 seconds.

The amount of pressure applied to the powder also gov-

erns the characteristics obtained in the final product. If the pressure is too low there will be insufficient adherence of the particles and the product will fracture; excessive pressure, however, will give good structural strength but the powder may be cold worked to such an extent that extreme grain growth will occur. Entrapped air in such a densified compact would give blowholes upon sintering. Elimination of the effects of various pressures was made by using the same pressure for each compact. The pressure used was 102,900 pounds per square inch; this pressure was concluded to be sufficient since the compacts were tightly bonded. A Rockwell hardness of R_f 42 was obtained from the compacts.

Returning to the equilibrium diagram of the Cu-Mn system shown below in Figure 1, it is seen that a solid solution

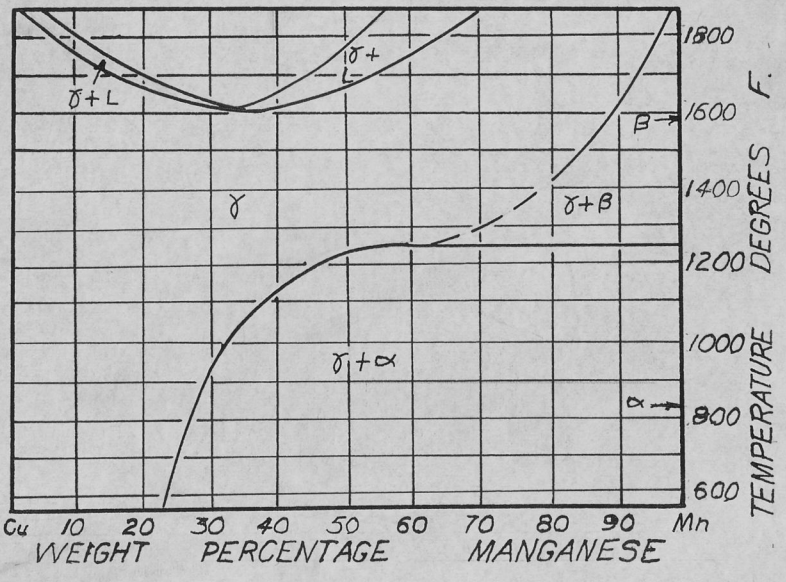

Figure 1

of Mn in Cu exists over a large range. Considering only a mixture of 60% pure Cu and 40% pure Mn it is seen that above 1150°F the Mn and Cu exist as a single phase δ solid solution. This composition is definitely age-hardenable; and it was decided that this approximate percentage would be ideally suited to the investigation.

Originally, the intention was to observe the possibilities of age-hardening Cu rich, Cu-MnS mixtures. It was immediately seen, however, that there was no diffusion occurring in these mixtures, even when heated to 1900°F which is only approximately 80 degrees below the melting of Cu. This lack of diffusability of Mn or MnS was attributed to the high affinity of Mn, over Cu, for S. This approach was then discontinued in favor of the elemental mixtures of Cu, Mn and S.

Since so little previous work had been done in the Cu-Mn-S system and none at all with the system as powders, the task was approached rather blindly. There was only a vague conception of possible results and each step was made with the idea of finding out as much as possible about the system from microstructural studies.

EQUIPMENT

The die used to compact the samples was made in the Chemical Engineering shop. A 2 inch steel (S.A.E. 5200) rod was turned down to a 1 3/4 inch diameter and a 0.25 inch center hole was drilled. This was then heat treated at 1600°F for 1½ hours, water quenched, and tempered 1 hour at 600°F. The center hole was then ground to 0.2538 inch diameter with a rotating burr. Both a base plug and top plug were ground from a tool steel rod, that had previously been heat treated, to give a clearance of 0.00025 inches. The die and one of the compacts is shown in Figure 2, below.



Figure 2. Exploded view of die assembly showing sample compact between base and top plugs.

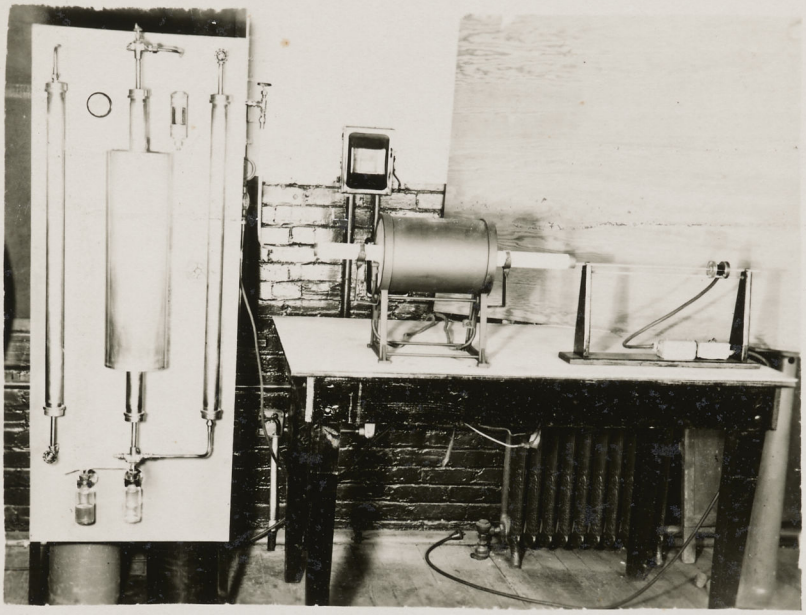

Figure 3. Nitrogen purifier with furnace.

A Brinell Hardness Testing Machine, manufactured by the Detroit Testing Machine Co., Detroit, Michigan, Model HB-2, serial number 222, was used to apply the load of 102,900 pounds per square inch to each compact. This pressure was computed using a 3,000 kilogram load on a 0.2538 inch diameter ram.

The furnaces used for heat treatment were a Type B-8 220 volt A.C. or D.C., Burrell High Temperature Globar Furnace having a reducing atmosphere of hydrogen present; and Hoskins Electric Furnace, Type F.H.-305, serial number 17997, manufactured by the Hoskins Manufacturing Co., Detroit, Michigan, utilizing an inert atmosphere of purified nitrogen.

The nitrogen atmosphere was made oxygen and moisture free by passing it through a series of three chambers containing drierite, heated Cu gauze and anhydrous magnesium perchlorate, respectively. The apparatus was constructed by J. Woods for use in previous metallurgical work. The purifier is shown in Figure 3.

After sintering the samples were mounted in Lucite using a small Fischer hydraulic press at a pressure of 2,000 pounds per square inch and a temperature of 260°F.

The samples were then taken through a series of polishing operations. They were as follows:

(1) Rough finish obtained from a Porter-Cable belt grinder, Type W-64, manufactured by the Porter-Cable Co., Syracuse, New York.

(2) Smooth finish obtained from use of a series of polishing papers having 1/0, 2/0, and 3/0 quality gradings. The paper used was Behr-Manning Emery Polishing Paper.

(3) Final high polish was obtained using a wet rotating wheel covered with Selvyt polishing cloth and levigated alumina.

The photomicrographs of the polished samples were made on a Bausch and Lomb Metallograph, serial number BR6555, manufactured by the Bausch and Lomb Optical Co., Rochester, New York.

EXPERIMENTAL PROCEDURE

Manganese sulfide was prepared by heating a mixture of sulfur, manganous sulfate and potassium carbonate in a ratio of 1 : 12 : 12[1]. The mixture was heated to the fusion point in a crucible over a bunsen burner with the evolution of sulfur dioxide. When the evolution ceased and the mass became a dull red it was allowed to cool and the soluble salts leached out with water in a vacuum filter. The resulting solid mass was a green powder.

The green manganese sulfide was dried in an atmosphere of hydrogen in a large test tube arranged with a two hole rubber stopper. A long glass tube, which reached almost to the bottom of the test tube was inserted in the stopper and the outlet of the test tube was connected through a conventional liquid trap to indicate the passage of gas. The entire test tube was inserted into a small upright furnace at a temperature high enough to vaporize any remaining sulfur in the sulfide. Hydrogen was passed through the equipment and the powder dried.

The dried powder was mixed with copper powder in a 3 to 97 weight ratio by tumbling in a glass jar attached to a variable speed stirring motor. Mixing was continued for approximately sixteen hours.

The compacts were made by use of a die mounted on a

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1. Meller; op. cit., page 388.

Brinell hardness tester using a total of 102,900 pounds per square inch pressure. After compacting, the samples were sintered in an inert or reducing atmosphere at varying temperatures. When the samples had cooled they were mounted in Lucite and polished.

Microscopic examination was accomplished on a large Bausch and Lomb metallograph. Photographs were also made on this metallograph.

When it was determined that the original manganese sulfide-copper system was producing no tangible results it was decided to use elemental manganese, copper, and sulfur. Electrolytic manganese was obtained from the Electromanganese Company of Knoxville and was powdered in a pebble mill. Varsol was used in the mill to prevent an explosion since powdered manganese is very pyrophoric in air. Ether was used to separate the powder from the remaining large pieces and to dissolve away the waxy residue of Varsol. After filtering off the ether and drying, the powder was screened through a series of Tyler screens; and only that passing a 350 mesh screen was used.

Copper, manganese and sulfur were mixed in a 57 to 40 to 3 ratio in the tumbler described above. Compacts were made of the mixture and sintered at varying temperatures. Some of the compacts were sintered overnight at 350°C in an upright furnace in a nitrogen atmosphere.

A mixture of 40% manganese, 58.5% copper and 1.5% sul-

fur was made and treated as were the others. Samples were sintered at 1200, 1300, 1470 and 1600°F for 15, 30, 45, and 60 minutes. These were mounted, polished and examined. All these samples were presintered at 350°C overnight.

A mixture of 60% copper and 40% manganese was prepared for comparison with the ones containing sulfur. In comparison, the samples were sintered for one hour at 1140°F and then 24 hours at 1479°F, then mounted, polished, examined, and photographed.

Samples sintered for one hour at 1300 and 1470°F were water quenched for comparison with the rest of these two series.

RESULTS AND DISCUSSION

The results obtained in this investigation are herewith presented in the form of photomicrographs along with a discussion of each picture or related series of pictures. The pictures are arranged in such an order as to give a conception of the progress of the investigation and the trends which occurred. The discussion is in regard to microstructure and the possible formation of solid solutions and sulfides.

No etching reagent could be found to bring out any contrast in the solid Cu grains without completely removing the dark gray matrix of the sample. Therefore all of the following photomicrographs are of unetched samples.

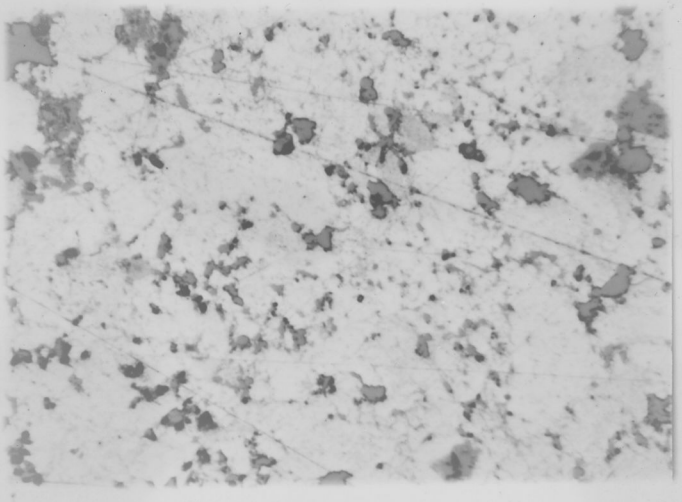

Figure 4. 97% Cu, 3% MnS. In as pressed condition[1].

1. All photomicrographs are at a magnification of 250x unless otherwise specified.

From the above photomicrograph it was apparent that mixing was thorough, as was illustrated by the uniform distribution of the small dark areas of MnS. The small black areas were holes in the sample; these were attributed to the fact that particles of MnS were pulled out during polishing. These loose particles also account for the numerous scratches that appeared on the surface.

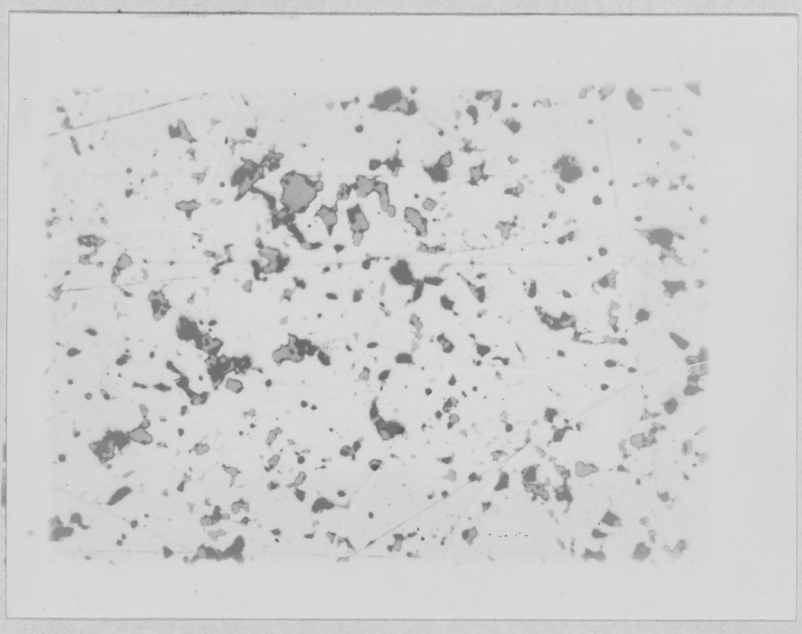

Figure 5. 97-3[1]. Sintered at 1600°F for one hour.

As illustrated in Figure 5, the sintering process caused very little change in the appearance of the samples from the as pressed condition. The only change noted was the disappearance of the individual Cu particles, which apparently fused to form a more or less solid mass. This treatment

1. 97% Cu, 3% MnS

showed no evidence of diffusion of the MnS into either the Cu grains themselves or along the grain boundaries. No formation of inter-metallic compounds or dissociation of the MnS to form free elements occurred.

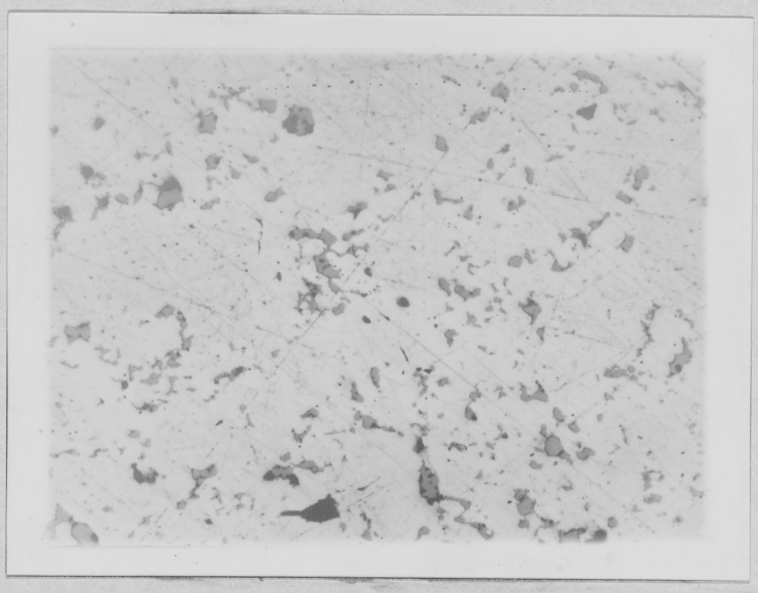

Figure 6. 97-3. Sintered at 1700°F for one hour.

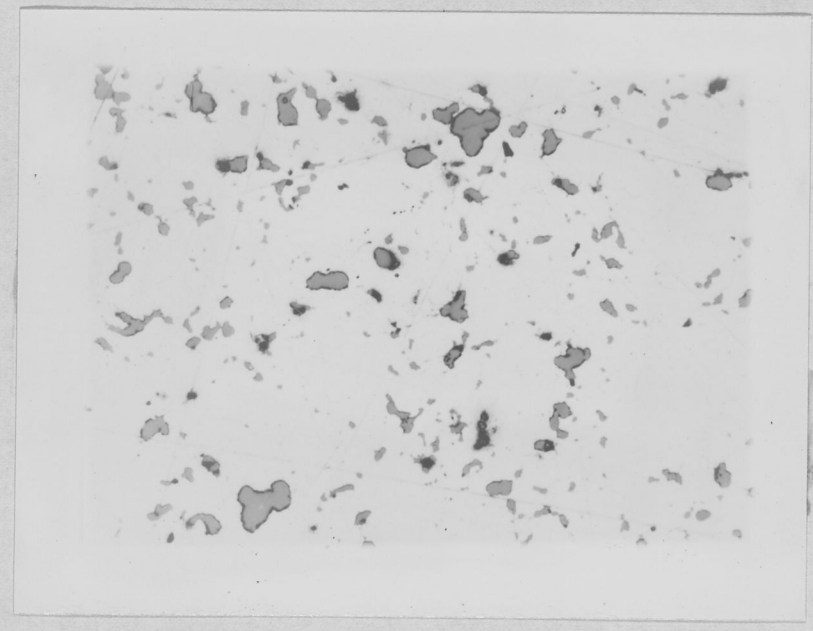

Figure 7. 97-3. Sintered at 1800°F for one hour.

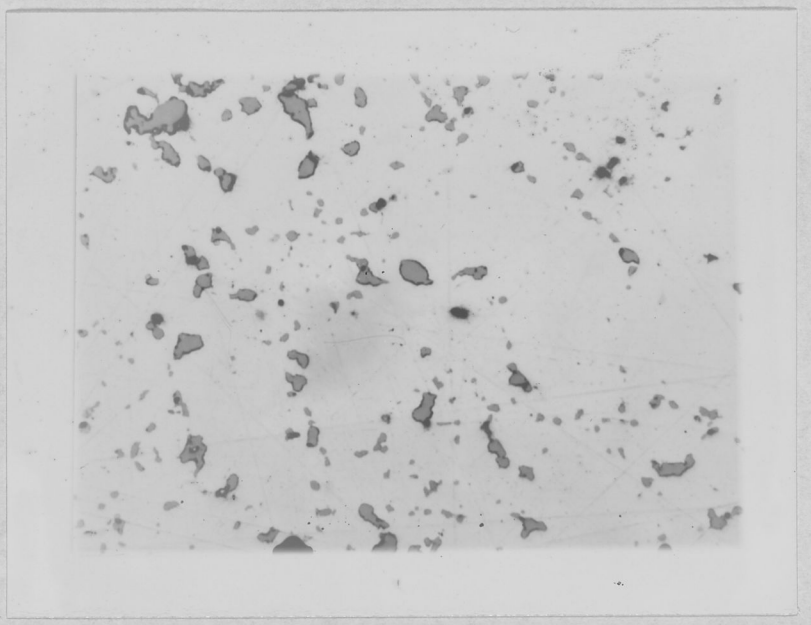

Figure 8. 97-3. Sintered at 1900°F for one hour.

Figures 6, 7, and 8 clearly illustrated that increased sintering temperature had no effect whatsoever upon the structure of the samples. Further increases in temperature were inadvisable since the melting point of Cu is 1981°F; and at 1900°F there was evidence of some melting occurring within the samples.

Since a solid solution of Mn in Cu are obtainable in this process only by diffusion and since there was no diffusion in the Cu-MnS system, this approach was discontinued. From the phase diagram of the Cu-Mn system it was noted that a definite solid solution of Mn in Cu existed. This being so, the approach was modified to start with the elemental Cu, Mn and S.

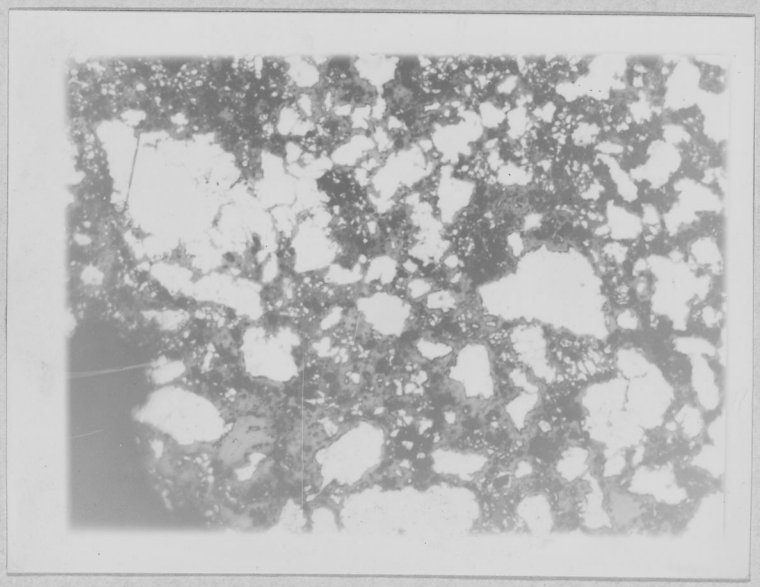

Figure 9. 57-40-3[1]. Sintered at 1140°F for one and one-half hours.

Examination of Figure 9 shows definite light gray areas around the majority of the Cu grains. These areas were thought to be non-metallic in nature since the sample had not been etched and they were much darker than a metallic solid solution would be assumed to be.

The sample, after polishing, exhibited a large number of cavities, some of which were extremely large in proportion to the majority of the cavities. The large black area in the corner of Figure 9 was a cavity attributed to the loss of a large sulfur particle either by volatization during sintering or by removal during polishing. Since it was

1. 57% Cu, 40% Mn and 3% S.

originally thought that a portion of the porosity might have been due to vaporization of entrapped moisture, a sample was weighed, dried in an inert atmosphere at 110°C, and re-weighed to determine any losses. There was no loss or gain in weight and it was concluded that no moisture was present.

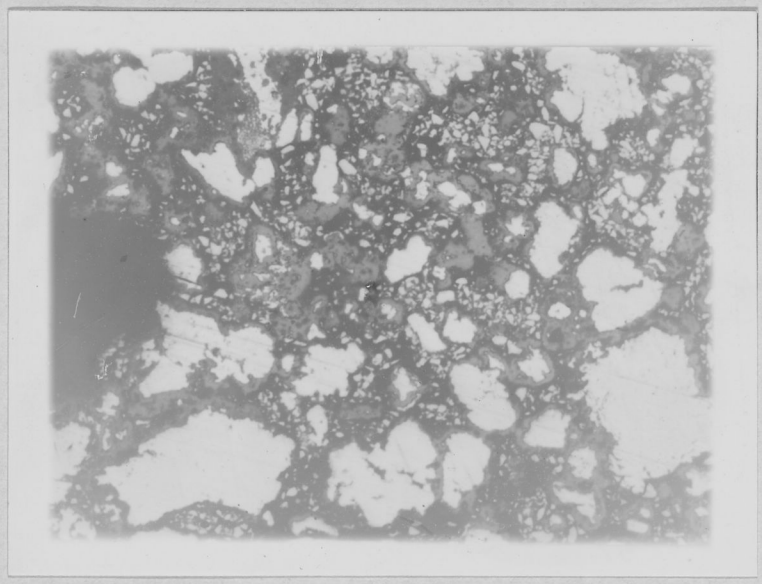

Figure 10. 57-40-3. Sintered at 1475°F for one hour.

In Figure 10 it was observed that the formation of the non-metallic substance was more pronounced, as illustrated by the increase in size of the light gray areas and the lacy appearance of some of the Cu grains. Increased sintering temperature had no effect, however, on the number of cavities.

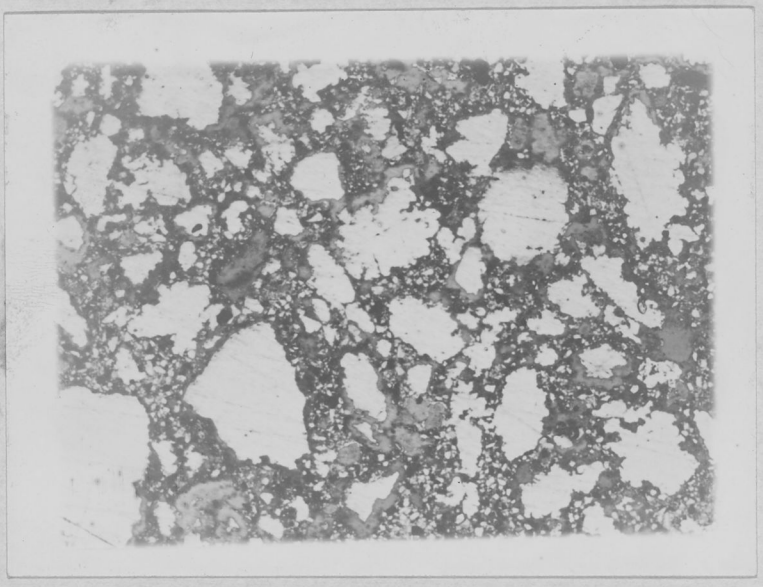

Figure 11. 57-40-3. Sintered overnight at 652°F and at 1470°F for one hour.

To determine whether or not the cavities were due to the condition of the sulfur, a sample was sintered for 22 hours at 652°F, which is in the liquid sulfur range. It was thought that by this procedure the sulfur would have time to react with the Cu or Mn if such reaction were to occur; and that in so doing any tendency of the sulfur to volatilize would be reduced. This was, indeed, the case and the number of cavities was found to be considerably smaller.

By comparing Figures 10 and 11 it may be seen that the extent of diffusion was approximately the same in both cases.

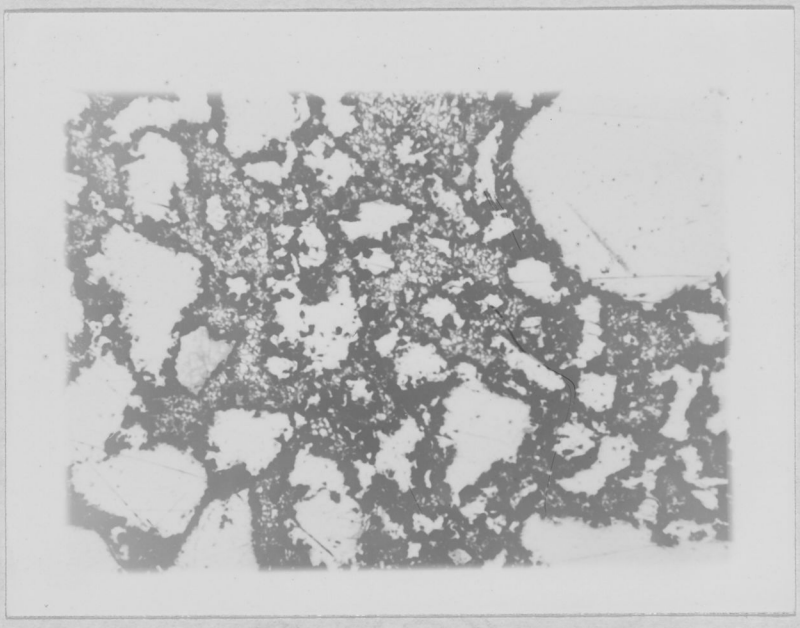

Figure 12. 60-40. Sample in as pressed condition.

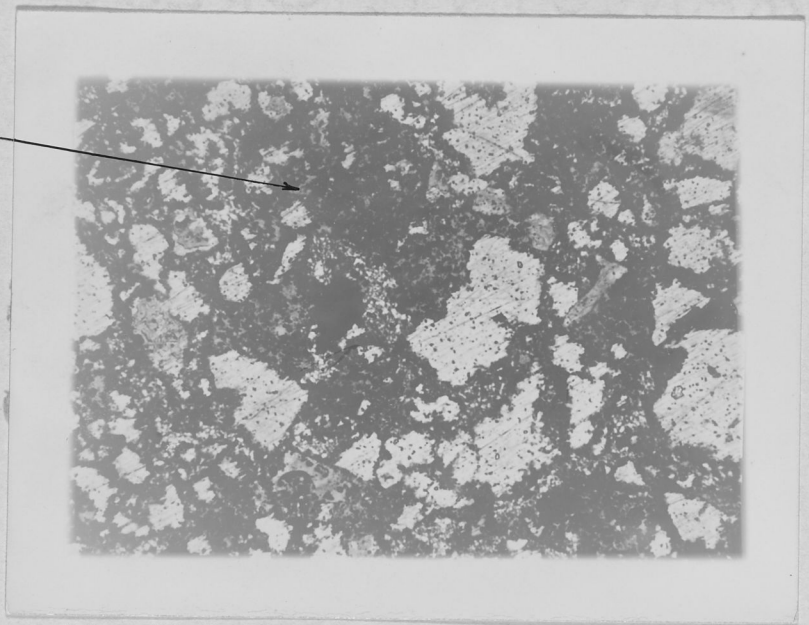

Figure 13. 58.5-40-1.5. Sample in as pressed condition.

Figures 12 and 13 showed the relative structures of two samples in the as pressed condition. This comparison was made to determine the original distribution of the S in Figure 13. The finely dispersed dark gray areas were thought to be S. The great abundance of the element in relation to its actual weight percentage was due to its large bulk in comparison with the other two elements present.

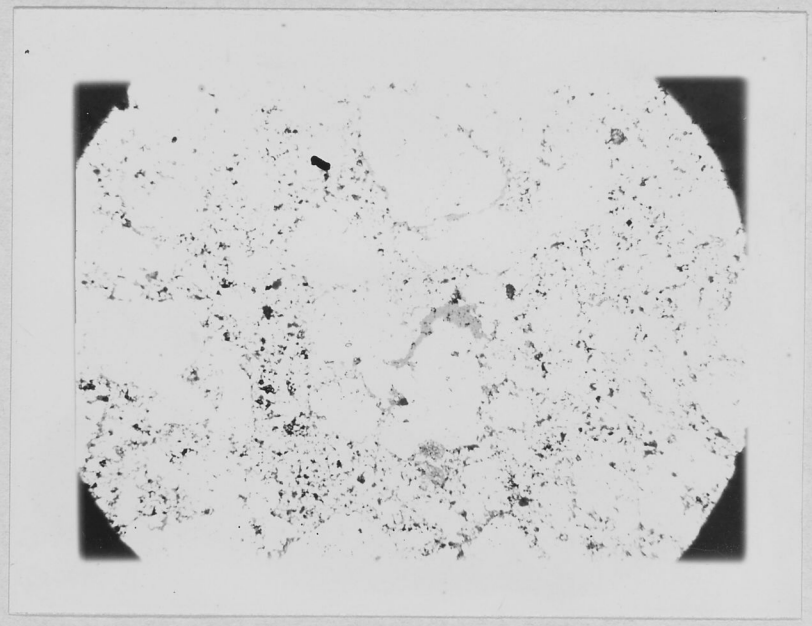

Figure 14. 60-40. Sintered at 1140°F for one hour and 1470°F for 24 hours. Magnification 150x.

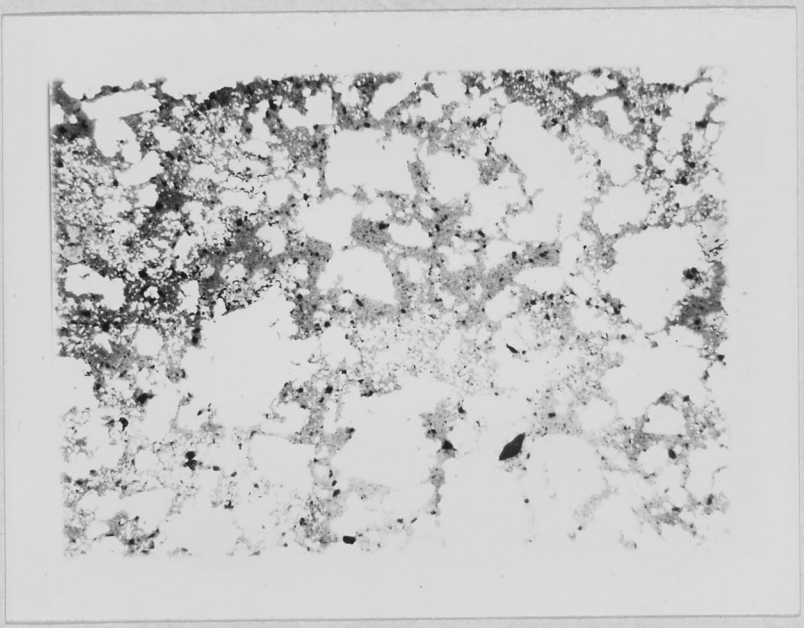

Figure 15. 58.5-40-1.5. Sintered at 1140°F for one hour and 1470°F for 24 hours.

As shown by Figures 14 and 15, the presence of a small amount of S apparently formed a large volume of sulfides, as was noted by the increased proportion of the gray areas in Figure 15 over those in Figure 14. These sulfides may either have been MnS, Cu_2S, a mixture of the two or a Cu-Mn-S complex. The black areas shown in both Figure 14 and Figure 15 were probably uncombined Mn particles. It was also noted that the reduction of the percentage of S from 3% to 1.5% completely eliminated the formation of large cavities. Accordingly it was decided that further investigation of this system was advisable.

The investigation was approached from the standpoint of a time-temperature relationship. The samples were sintered at temperatures of 1200, 1300, 1470, and 1600°F and for 15, 30, 45, and 60 minutes at each temperature.

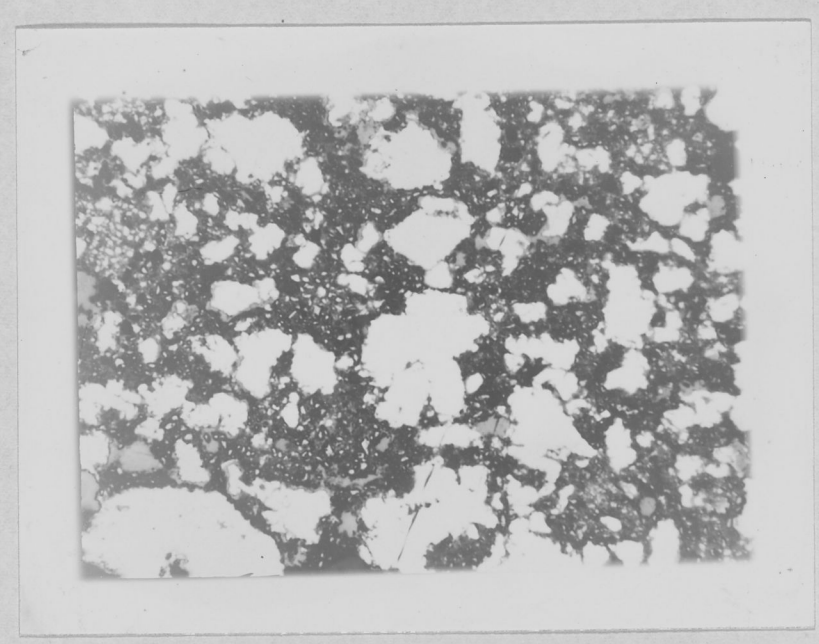

Figure 16. 58.5-40-1.5. Sintered at 1200°F for 15 minutes.



Figure 17. 58.5-40-1.5. Sintered at 1200°F for 30 minutes.

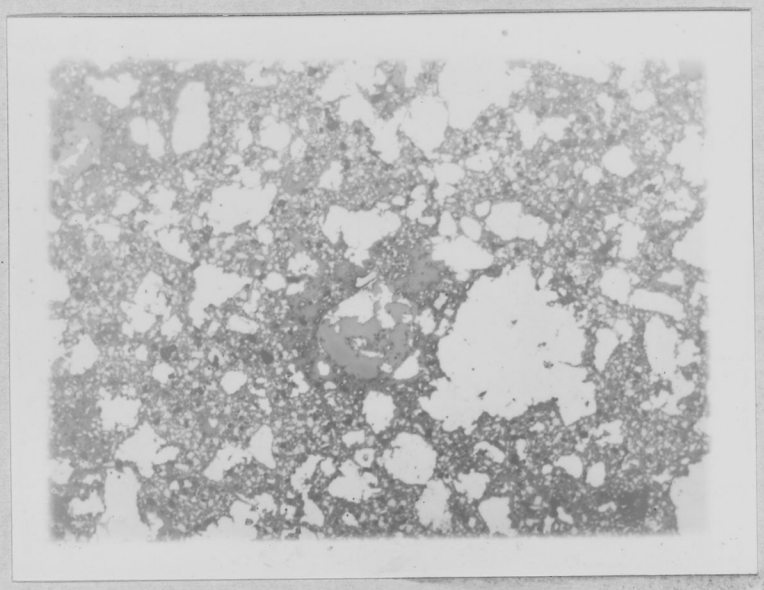

Figure 18. 58.5-40-1.5. Sintered at 1200°F for 45 minutes.

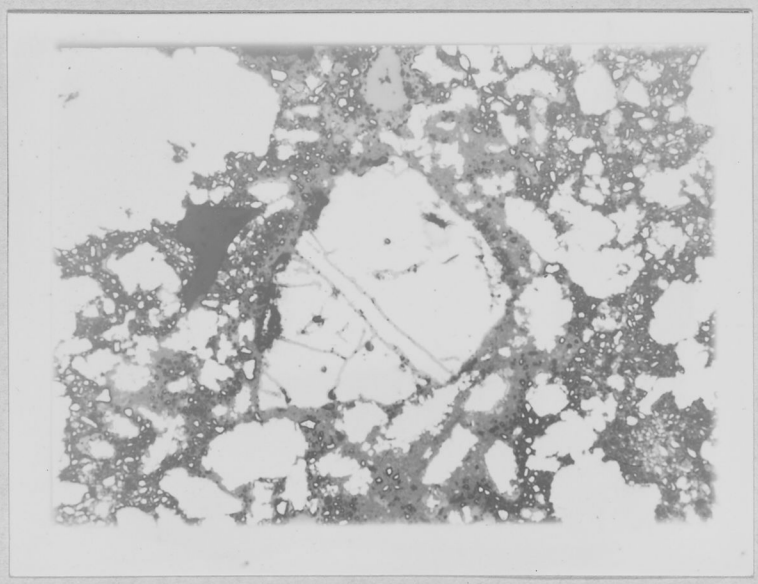

Figure 19. 58.5-40-1.5. Sintered at 1200°F for 60 minutes.

Sintering at 1200°F for 15 minutes showed only slight formation of the gray areas previously identified as sulfides of either Mn, Cu or both. This is illustrated in Figure 16. By further treatment at 1200°F for times of 30, 45 and 60 minutes the results shown in Figures 17, 18 and 19 were obtained. It was noted from these photomicrographs that increased sintering time increased both the amount of sulfides present and the diffusion of the sulfides into the Cu grains themselves.

If it is assumed that the sulfide formed is Cu_2S then the maximum sulfide that could be present is about 7.5% by weight of the entire structure. There is definitely a much greater amount of dark appearing constituent present. Since MnS would even a smaller area than Cu_2S it seems that the dark matrix could not be either Cu_2S, MnS or a mixture of the two sulfides but rather, perhaps, a complex sulfide of Cu and Mn. With S exhibiting its maximum valence of seven and the Cu and Mn their lowest valences then a sulfide might be formed utilizing all the S and forming the dark matrix. The completely black areas are probably accountable for as uncombined Mn.

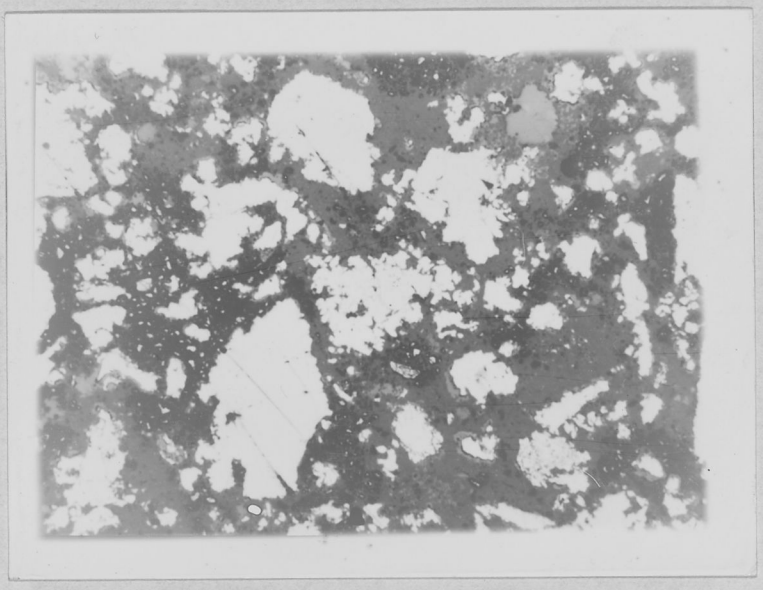

Figure 20. 58.5-40-1.5. Sintered at 1300°F for 15 minutes.



Figure 21. 58.5-40-1.5. Sintered at 1300°F for 30 minutes.

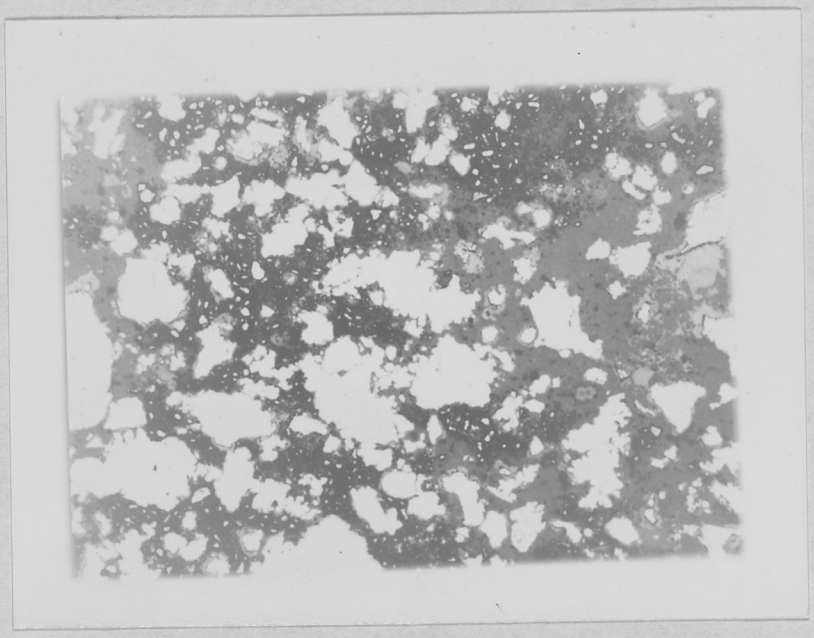

Figure 22. 58.5-40-1.5. Sintered at 1300°F for 45 minutes.

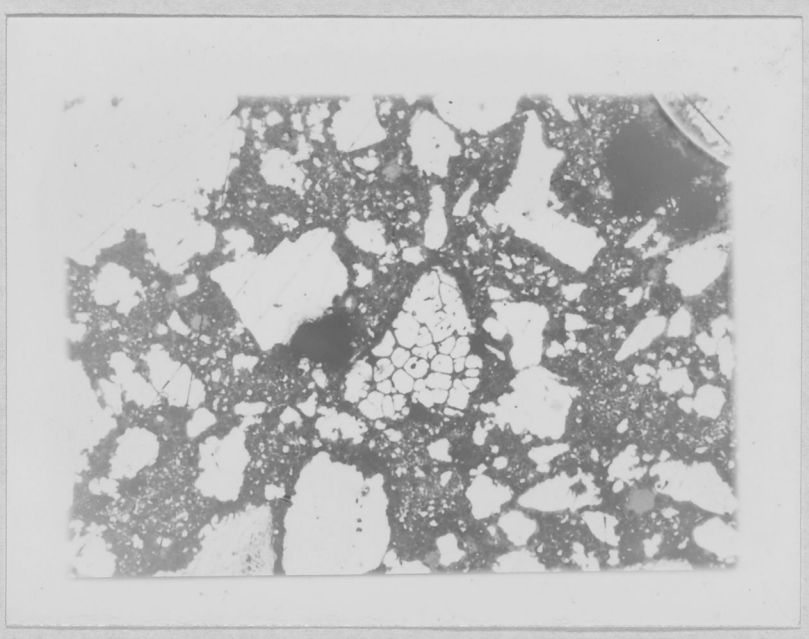

Figure 23. 58.5-40-1.5. Sintered at 1300°F for 60 minutes.

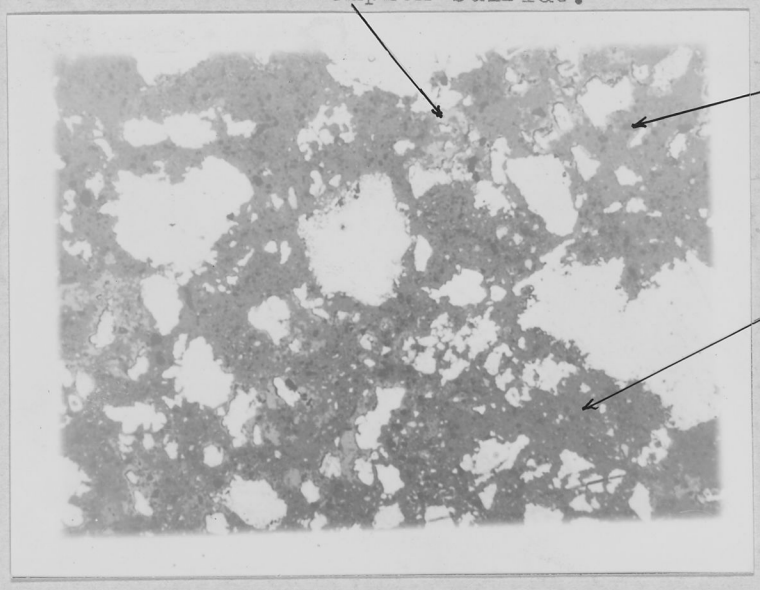

Figure 24. 58.5-40-1.5. Sintered at 1300°F for 60 minutes and then water quenched.

The series sintered at 1300°F showed the same general trends as did the 1200°F series. The only differences noted were the larger amounts of sulfides and increased amount of diffusion at 1300°F. In addition, one sample was water quenched after one hour at 1300°F, and a comparison between Figures 23 and 24 showed a decided increase in the amount of sulfides present in the quenched sample over that of the slow cooled samples. This difference indicated that definite precipitate occurred on slow cooling thereby reducing the amount of sulfides visible.

As may be seen from examination of the Cu-Mn phase diagram, 1300°F is well into the range of the α solid solution of Mn in Cu. It is entirely possible that this solution would exist at this sintering temperature. This solid solu-

tion might possibly react with the sulfur or with any sulfides formed to add to the gray areas. There are some lighter gray areas adjacent to some of the Cu grains as indicated in Figure 24. These might be such a solid solution complex sulfide. The preponderance of gray matrix in Figure 24 over Figure 23 and the finely dispersed particles in Figure 23 seemed to indicate, also, that this might be true. Any solid solution would remain so upon quenching and would precipitate out Mn on slow cooling.

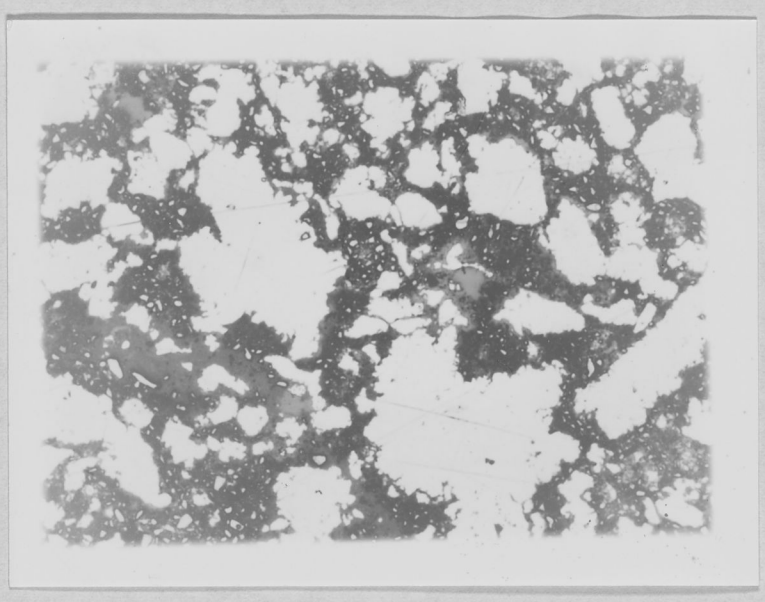

Figure 25. 58.5-40-1.5. Sintered at 1470°F for 15 minutes.

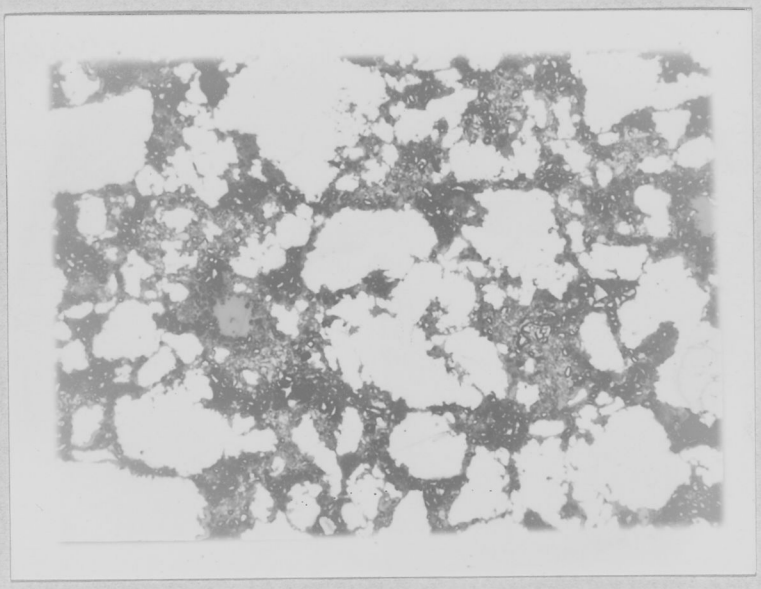

Figure 26. 58.5-40-1.5. Sintered at 1470°F for 30 minutes.

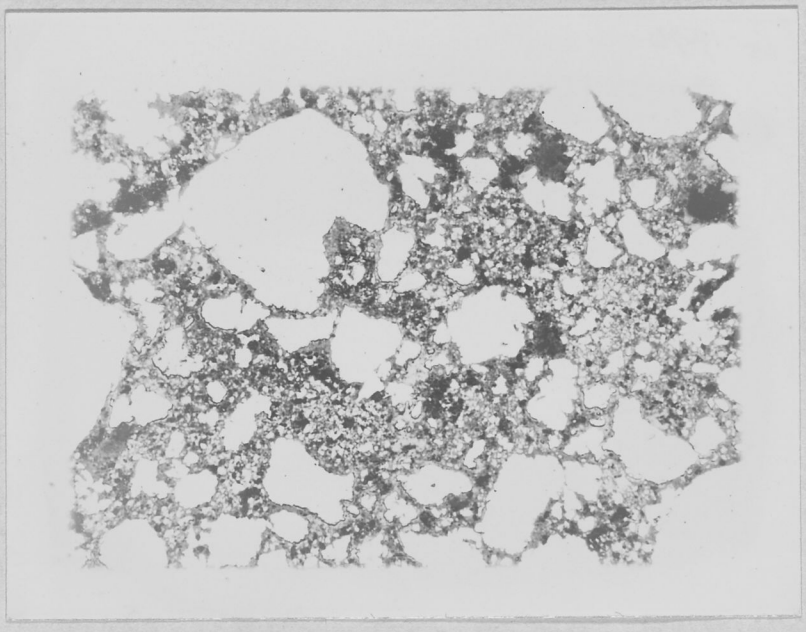

Figure 27. 58.5-40-1.5. Sintered at 1470°F for 45 minutes.

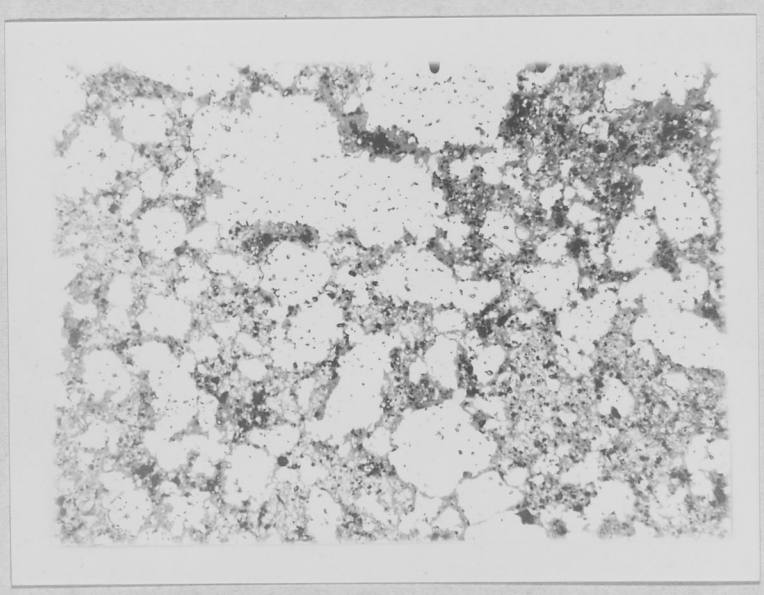

Figure 28. 58.5-40-1.5. Sintered at 1470°F for 60 minutes.



Figure 29. 58.5-40-1.5. Sintered at 1470°F for 60 minutes and water quenched.

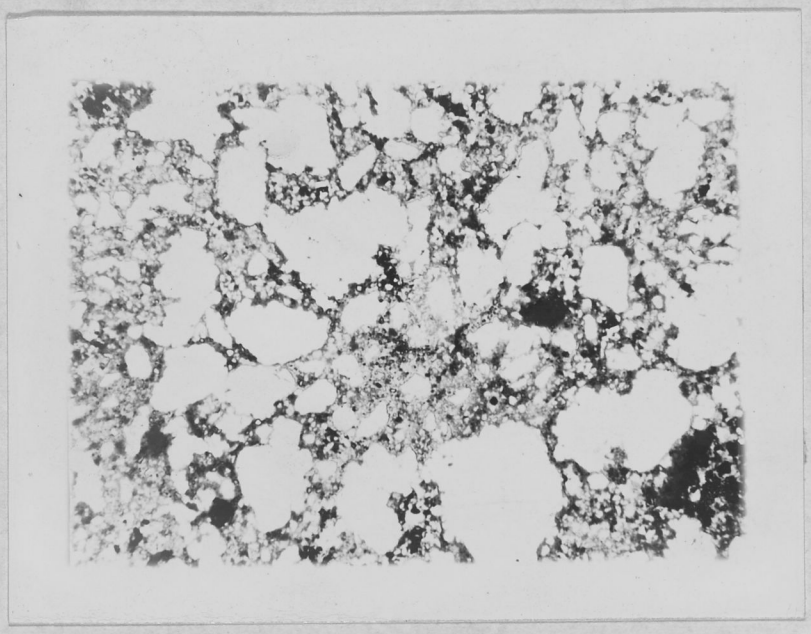

Figure 30. 58.5-40-1.5. Not sintered overnight. Sintered at 1470°F for one hour.

For some reason the series sintered at 1470°F did not show the expected trends in the same manner as did the previous two series. Instead of an increased amount of visible sulfide, a finely divided precipitate occurred throughout the matrix. The amount of this precipitate increased with increasing time as shown in Figures 25, 26, 27 and 28. Figure 29, however, seems to indicate that a complex sulfide was formed and in large amounts. Since this precipitation occurred it indicates that the dark gray areas formed may have been solid solution and the fine constituent precipitated Mn. Further proof that the fine constituent in the matrix of Figure 28 was precipitated is shown in the fact that a similarly sintered sample, when quenched, exhibited none of the precipitate but a fairly smooth gray matrix. See Figure 29.

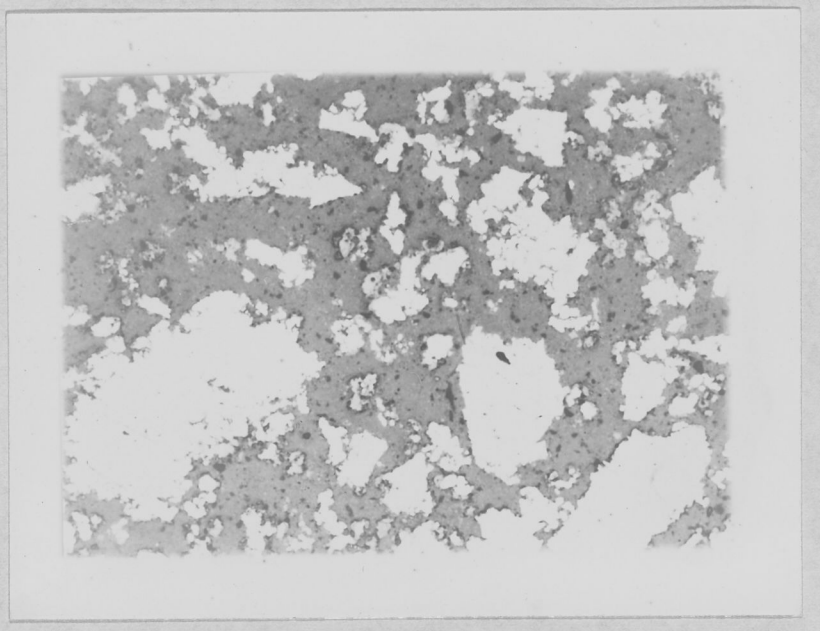

Figure 31. 58.5-40-1.5. Sintered at 1600°F for 15 minutes.

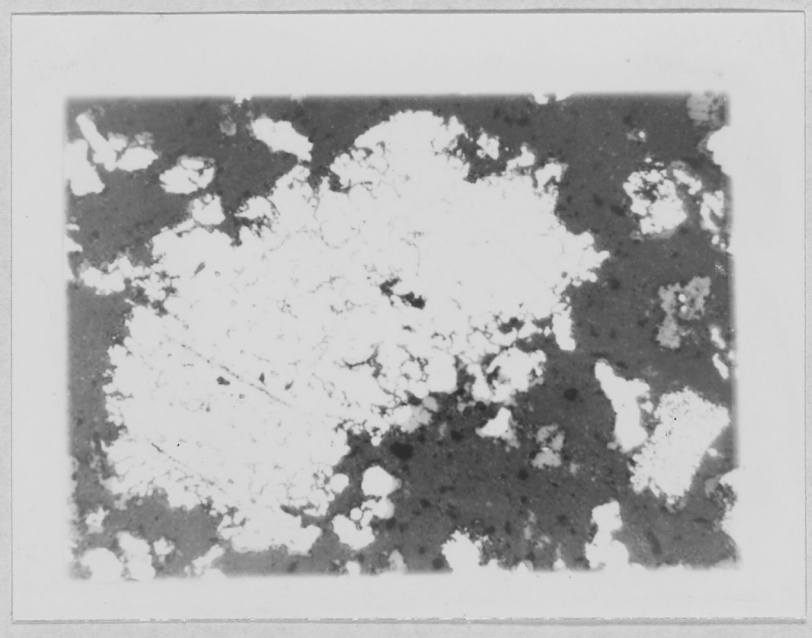

Figure 32. 58.5-40-1.5. Same sample as above.

Magnification 450x.



Figure 33. 58.5-40-1.5. Sintered at 1600°F for 30 minutes.



Figure 34. 58.5-40-1.5. Sintered at 1600°F for 45 minutes.

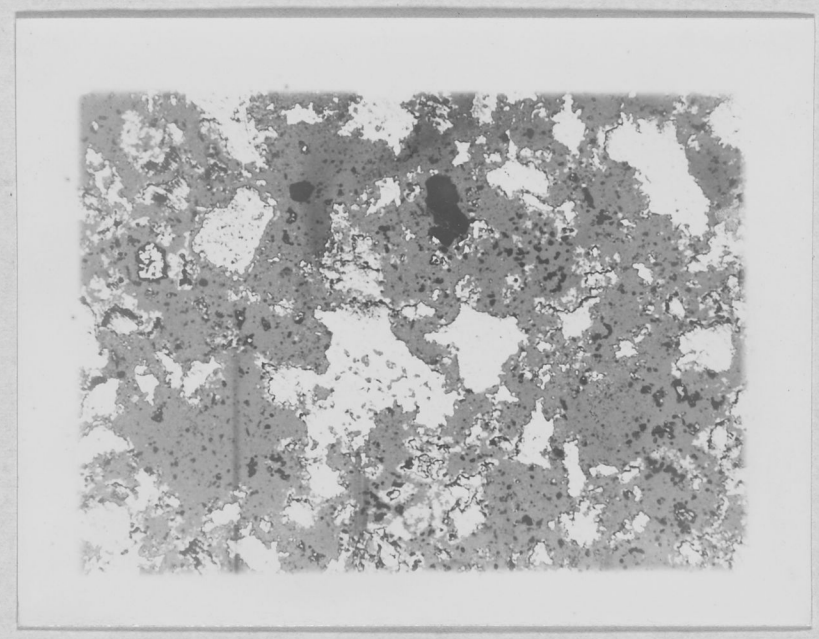

Figure 35. 58.5-40-1.5. Sintered at 1600°F for 60 minutes.

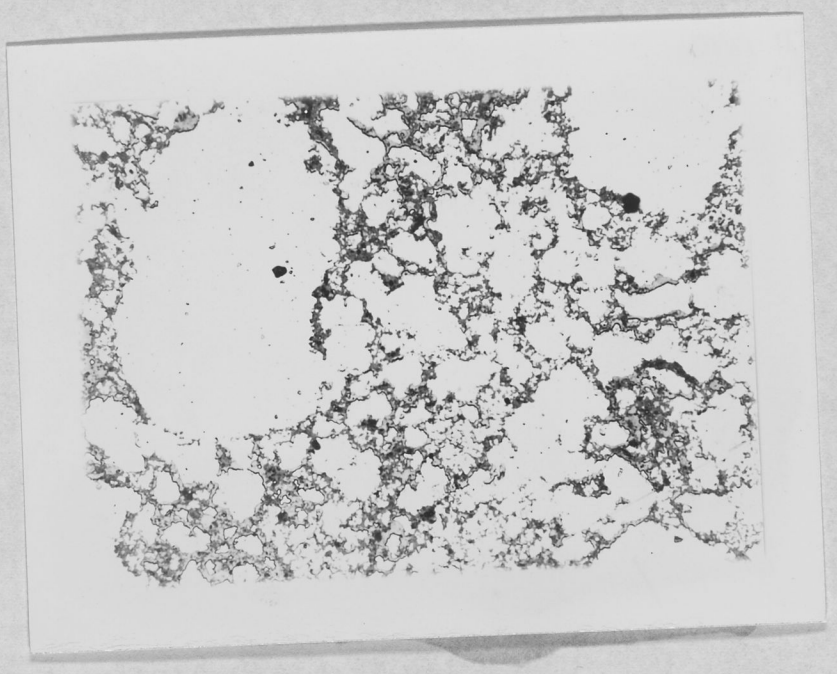

Figure 36. 58.5-40-1.5. Sintered at 1600°F for 60 minutes.

Sample not presintered.

The appearances of the structures of this series are quite different from those of the 1470°F series. Apparently almost complete reaction took place between the large Cu grains in fifteen minutes. A great deal of diffusion into the Cu grains took place in that time as indicated by Figure 32. Increased sintering time served only to increase the diffusion into the larger grains. Many of the large grains that appear to be almost completely consumed could possibly be a cluster of small Cu particles that had been pressed together on compacting.

A very interesting phenomenon may be observed by comparing Figures 35 and 36. The sample illustrated in Figure 36, not having been presintered, showed very little sulfide formation. It was thought that this result was due to a lack of reaction between Mn and S. The presintering of the samples in the liquid S range would serve to allow the S to flow around the Mn and Cu grains and come in very intimate contact with them. This would increase the rapidity with which a reaction between constituents would take place. Any reaction which took place must have been rapid since so much gray area was produced in fifteen minutes at 1600°F. The lack of formation of large amounts of the complex sulfides, as shown in Figure 36, could have been due to volatilization of the S from the compacts on rapid heating of the compacts to 1600°F.

CONCLUSIONS AND RECOMMENDATIONS

The results of this investigation are by no means completely conclusive. They appear to only scratch the surface of a field exhibiting a great number of possibilities for future investigation. The lack of knowledge of this system made very difficult to obtain conclusive results since there were no previous data with which to work.

The negative results obtained using MnS and Cu compacts may have been due to several things. First, the free energy of MnS is -54,000 cal. per mole of S_2 while the free energy of Cu_2S is -49,000 cal. per mole of S_2[1]; second, MnS is higher in the electrochemical series than is Cu_2S and would tend to remain as MnS[2]. For these reasons many investigators feel that Mn has a greater affinity for S than does Cu. It would appear that this greater affinity does exist since no reaction was discernible in the MnS-Cu compacts. It is recommended that investigation be made of this system using much larger percentages (30-50%) of MnS.

Under the conditions used in this investigation it was found that a S content above approximately 2% caused a large number of cavities to be formed in the sintered samples.

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1. "The Influences of Temperature on the Affinity of Sulfur for Copper, Manganese and Iron." E. M. Cox, M. C. Bachelder, N. H. Nachtrieb and A. S. Skapski; Journal of Metals Technology and Practice, January, 1949, pages 27-31.
2. Meller; op. cit., page 397.

All samples containing 1.5% S showed a complete absence of cavities, while those containing 3% S contained a great number of cavities. These results, relative to each other, seemed to definitely establish that, under a given sintering procedure, a critical S content exists as regards cavity formation.

As the major portion of this investigation was based on 58.5% Cu, 40% Mn and 1.5% S, the following conclusions and recommendations refer only to this system.

In pursuing this investigation the foremost problem presenting itself was identification of the dark gray areas surrounding the Cu grains. These gray areas first began to appear only as a fringe around the larger Cu grains. With increases in sintering temperature these areas increased proportionately. They were thought to a Mn rich complex sulfide of Mn and Cu. Also in evidence were some light gray areas which occurred either adjacent to or surrounding a Cu grain. Due to their proximity to the Cu grains they were identified as probably being Cu rich complex sulfides of Mn and Cu. In some samples, such as Figure 35, there appeared areas which were first thought to be holes. Upon close observation it was seen that these areas were in the plane of the sample surface and were solid matter within the sample. No speculation was made as to the composition of the dark spots as regards the materials known to be present. These may have been foreign particles. It is recom-

mended that further investigation be made regarding accurate identification of the microconstituents.

By comparison of Figures 31 and 32, which are of the same sample at different magnifications, it may be seen that there is a great difference in color shades of the various constituents. These differences are due to photographic technique and could be eliminated by very careful and uniform exposure of plates and development of negatives and prints.

As an aid to identification of microconstituents, it is recommended that, in further investigations, examinations be made of compacts known to contain MnS or Cu_2S exclusively. These could be compared to compacts thought to contain either or both.

Since sulfides were apparently formed and since research on the materials was undertaken to determine the possibility of their use as bearing materials, it is recommended that further investigations containing a series of mechanical tests of the bearing qualities be made on these materials.

APPROVAL

The foregoing thesis is hereby approved as a creditable study of an engineering subject, carried out and presented in a manner sufficiently satisfactory to warrant its acceptance as a prerequisite to the degree for which it has been submitted. It is to be understood that by this approval the undersigned does not necessarily endorse or approve any statement made, opinions expressed, or conclusions drawn therein, but approves the thesis only for the purpose for which it is submitted.

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Date June 3, 1949