Vapor Phase Brazing of Copper Plated Carbon Steel in a Zinc Vapor

Vaporized zinc reacts with copper on copper-plated steel to form a molten brazing filler metal which flows into joint regions being brazed

BY T. OSAWA

ABSTRACT. A new brazing process, principle of which is based on the vapor phase reaction between the base metal and the metal vapor, was studied from a metallurgical viewpoint. Tests were undertaken to make copper-plated carbon steel react with zinc vapor in a protective atmosphere, in order to braze the carbon steel with "brass filler metal" that would be formed by vapor phase reaction of copper plating with zinc.

When using vapor phase brazing on carbon steel, the brazing temperature was lower than that for copper brazing and the brazement was uniformly coated by the gold-colored Cu-Zn system alloy. The zinc content of Cu-Zn alloy obtained in this process was affected by brazing temperature, and metallurgical structures at 1173, 1193 and 1213 K (1652, 1688 and 1724° F) were determined to be α phase, $(\alpha + \beta)$ phase and β phase in the Cu-Zn equilibrium diagram, respectively. The fillets formed at brazed joints by the process were faultless and by no means inferior to those obtained using ordinary brazing methods. The flow factor, evaluating brazeability, was at the range of 0.7 to 0.9, and superior brazeability was observed.

Introduction

Various brazing processes have been developed to a high degree of technical proficiency, and new brazing methods such as those utilizing a vacuum or protective atmosphere were evolved to join the difficult-to-braze base metals with tenacious oxide films (Ref. 1, 2). These methods use the same principle for brazing—namely, that low melting alloys as the brazing filler metals are made to melt and to flow into the joint gap (Ref. 3).

Work by the author has led to the development of a new brazing process whose principle differs from that of ordinary brazing methods. The process is based on vapor phase reaction between the base metals being brazed and the metal vapor being used as a brazing atmosphere—that is to say, low melting alloys, formed by the diffusion reaction between the base metals or pre-coated metals on the base metals and the metal vapor, are made to act as "filler metals". With this process, alloys formed by vapor phase reaction may act as a filler metal to flow and to form the fillets at the joints, if the alloys have lower melting points than the base metals.

In conventional brazing, it is essential that the filler metals have lower melting points than the base metals. On the other hand, in the vapor phase brazing process it is not necessary that the pre-coated metal on the base metal have a lower melting point than that of the base metal. What is necessary is that the filler metal have a lower melting point than the base metal when it is formed by diffusion reaction between the pre-coated metal and the metal vapor in a protective atmosphere.

In this study tests were undertaken to braze carbon steel with Cu-Zn system "filler metals" formed by vapor phase reaction between the copper plating on carbon steel and the zinc vapor in hydrogen atmosphere. The brazeability and the metallurgical microstructures of brazed interfaces obtained with this process were discussed in relation to brazing condition.

Figure 1 compares vapor phase brazing with the ordinary process.

Experimental Procedure

Material and Apparatus

Medium carbon steel plates (0.35% carbon content) with a 10 μm thick coating of electrically plated copper were prepared as the base metals. The base metals were degreased with the trichloroethylene and heated in the zinc vapor current at the temperature range of 1133 to 1213 K (1580 to 1724° F). At an elevated temperature the copper plating on the carbon steel plates may react with the zinc vapor to form a Cu-Zn system alloy to flow into the joints, if the alloys are in a molten state. The zinc vapor was produced by means of heating the grainy zinc in a hydrogen atmosphere controlled at the same temperature as the brazing temperature.

Figure 2 is a schematic drawing of the experimental apparatus for the vapor phase brazing. Main components are a fused silica tube, a stainless steel bar for

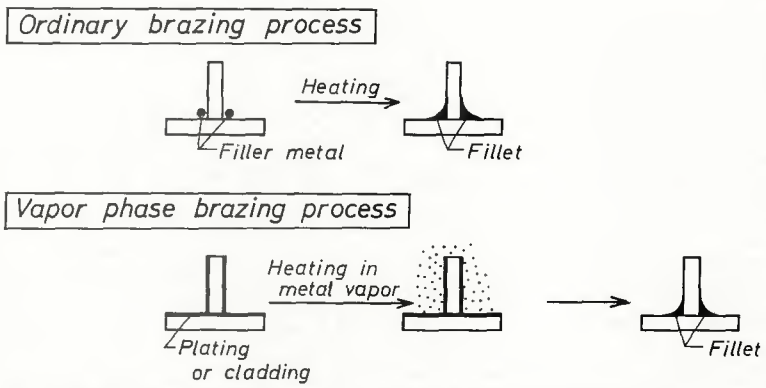

Fig. 1—Schematic comparing vapor phase brazing to ordinary process

T. OSAWA is Lecturer, Faculty of Science and Engineering, Aoyama-Gakuin University, Tokyo, Japan

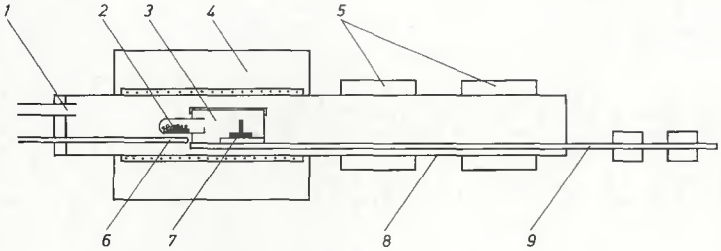

Fig. 2 — Schematic of experimental apparatus used for vapor phase brazing: 1 — hydrogen inlet; 2 — grainy zinc; 3 — stainless steel container box; 4 — electric furnace; 5 — cold trap; 6 — thermocouple; 7 — specimen; 8 — fused silica tube; 9 — stainless steel bar for inserting specimen

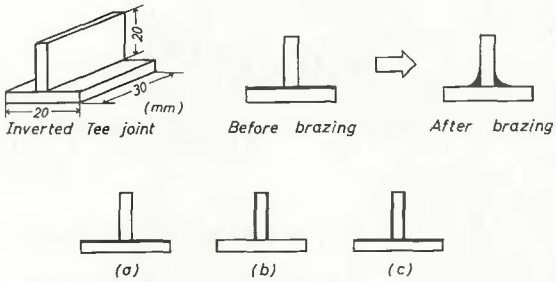

Fig. 3 — Inverted T-joint test specimens before and after brazing, and three types of inverted T-joints. The copper-plated base metal is set as follows: (a) — on the horizontal side (H type); (b) — on the vertical side (V type); (c) — on both sides (W type).

inserting test specimens, and a stainless steel container box with a chamber for evaporating zinc. Brazing occurs in the stainless steel container box.

Experiment

In this study the following items were investigated:

1. The temperature dependence of the amount of a Cu-Zn system alloy formed by diffusion reaction between the copper plating on the carbon steel plate and the zinc vapor.
2. The observation of metallurgical microstructures of the brazed joints.
3. Analysis of brazed joint interfaces by EPMA (Electron Probe Micro Analysis).
4. Evaluation of joint brazeability.

The zinc content of a Cu-Zn system alloy was measured by the weight gain from before and after the reaction. The brazeability that is the most important factor for brazing was evaluated by the amounts of fillet volume formed on the inverted T-joint shown in Fig. 3. The volume of fillet formed (V′) can thus be expressed as some fraction of the volume of a Cu-Zn system alloy formed (V) on the base metal by the reaction of copper plating with zinc vapor, or:

$$V' = k\,V \qquad (1)$$

where k will be referred to as a "flow factor".

It follows that:

$$V' = A_f\,l = \frac{W}{d} \qquad (2)$$

where A_f is the cross sectional area of the fillet; l is its length; W is the weight of the obtained Cu-Zn alloy; d is the density of its alloy. It is considered that the larger k is, the better the brazeability; this is because k indicates a contribution of the molten Cu-Zn alloy for the fillet of the joint.

In this experiment the flow factor k was measured for three different types of specimen of the inverted T-joint shown in Fig. 3 — that is, the copper-plated carbon steel plate which takes part in the reaction with the zinc vapor is put on the vertical side to the bare carbon steel plate (V), on the horizontal side to the bare carbon steel plate (H) and on the both sides (W). These joints were named V type, H type and W type, respectively.

Results and Discussion

Reaction between Copper and Zinc Vapor

Figure 4 illustrates an example of the microstructure of the brazed joint interface formed by the vapor phase brazing process. As can be seen, the fillets formed at the joint are the same as that of the ordinary method using the brass filler metal and flux.

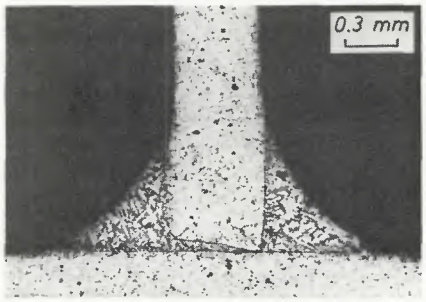

Fig. 4 — Example of brazed joint interface microstructure obtained with vapor phase brazing process

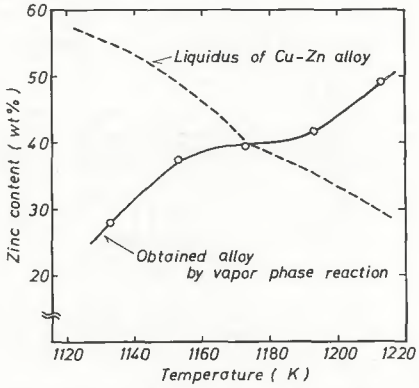

Fig. 5 — Relation between reaction temperature and zinc contents of the Cu-Zn system alloy obtained by vapor phase reaction of the copper plating (on the carbon steel) with the zinc vapor. The copper plating thickness is 10 μm, and the reaction time is 30 s

The reaction between the copper plating on the base metal and the zinc vapor was examined, because the reaction was particularly important in vapor phase brazing. Figure 5 shows the relation between the reaction temperature and the zinc contents of the Cu-Zn system alloy obtained by heating the copper plated carbon steel plate (copper plating thickness — 10 μm) in the zinc vapor at various temperatures for 30 seconds (s). It can be seen clearly that the formed Cu-Zn system alloy has a tendency to increase its zinc content with reaction temperature. This was proved by the weight gain shown after the reaction by the zinc content of the alloys; at 1133, 1173, and 1213 K (1580, 1652, and 1724° F). The weight gains were 27, 42 and 45%, respectively. Therefore, the metallurgical structures of these alloys were considered to be α, (α + β) and β phase in the Cu-Zn binary diagram (Ref. 4), respectively.

In Fig. 5 the dotted line shows the liquidus temperature quoted from the equilibrium diagram of a Cu-Zn alloy system (Ref. 4), and the full line shows the zinc content of the obtained alloys. Since the two lines intersect at about 1173 K (1652° F), the Cu-Zn alloy obtained at below 1173 K cannot contain as much zinc as does the alloy in liquid state and "brazing" seems impossible to perform. On the other hand, at the reaction temperature above 1173 K (1652° F), the specimen may be brazed to form the fillets on the joints because the formed Cu-Zn system alloy contains enough zinc to be in liquid state.

Figure 5 shows that the reaction time is 60 s; the zinc contents of the Cu-Zn system alloy may depend on the reaction time because the reaction of copper with zinc vapor is dependent on the diffusion reaction. In general, brazing time should be as short as possible, because longer brazing times cause undesirable effects such as the coarsening of base metal grains and the penetration of molten filler

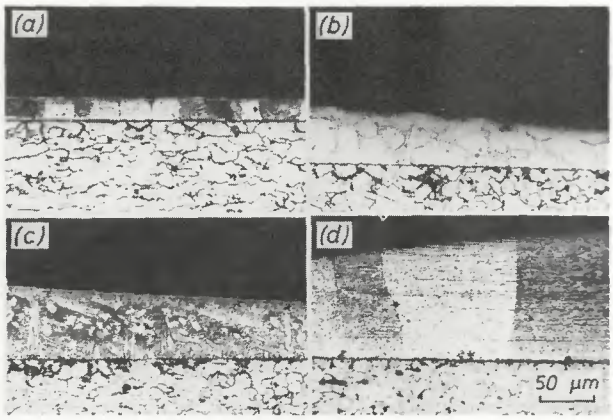

Fig. 6 — Microstructures of interfaces obtained at different vapor phase brazing temperatures: (a) — 1153 K (1616° F); (b) — 1173 K (1652° F); (c) — 1193 K (1688° F); (d) — 1213 K (1724° F)

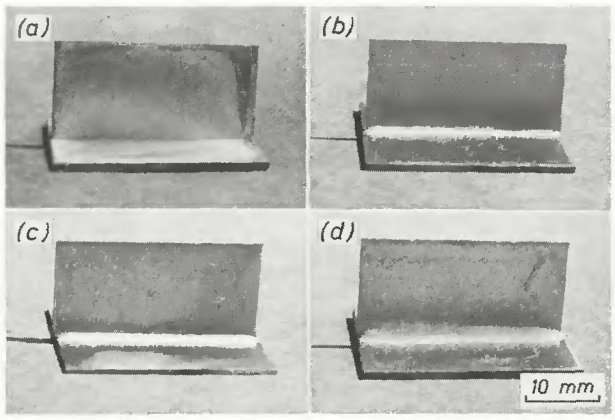

Fig. 8 — External appearance of various T-joint test pieces brazed with the vapor phase process at indicated temperatures: (a) — W type, 1153 K (1616° F); (b) — H type, 1193 K (1688° F); (c) — V type, 1193 K (1688° F); (d) — W type, 1193 K (1688° F)

metals into the grain boundaries (Ref. 5).

Figure 6 shows the metallographic microstructures of joint interfaces obtained by vapor phase brazing. As shown in Fig. 6, the metallurgical solidification structure cannot be observed for the specimen brazed at 1153 K (1616° F); therefore, the alloy seems not to have been in liquid state and the structure is mainly composed of α phase. On the other hand, the alloy formed at 1173 K (1652° F) must have been in liquid state since the structure showed the casting structure of α phase (although β phase was observed partially). Further, at 1193 K (1688° F) the structure shows the mixtures of $(\alpha + \beta)$ phases, and at 1212 K (1724° F) an homogeneous structure that appears to be β phase is observed.

Brazeability

Brazeability is generally evaluated by one or more methods (Ref. 6, 7, 8). In this investigation the flow factor (indicating the flowability of molten filler metal) was measured as a way to evaluate brazeability.

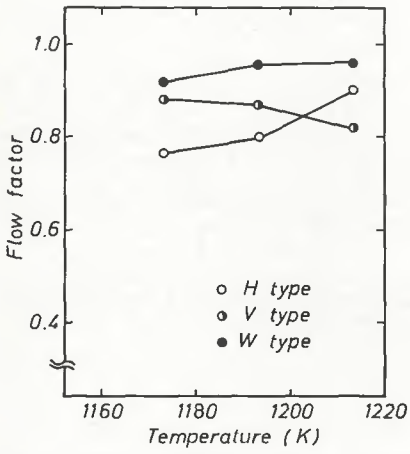

Fig. 7 — Effects of brazing temperature on the flow factors for various types of inverted T-joint specimens

Figure 7 shows the relationships between brazing temperature and flow factors for various types of inverted T-joints. It is obvious that the molten Cu-Zn alloy (i.e., filler metal) obtained by vapor phase brazing is so superior in the flowability that the Cu-Zn system alloy formed on the carbon steel base metal is almost entirely consumed in forming the joint fillet. This means that the flowability of the molten Cu-Zn system alloy is excellent. It is considered that the superior brazeability of this process may be due to the fewer interactions (such as diffusion of the molten alloy into the base metal, dissolution of the base metal into the molten alloy, etc.) between the carbon steel and the Cu-Zn system alloy; this is because the brazing temperature is lower when compared to the copper brazing.

As can be seen clearly from Fig. 7, the temperature dependence of flow factors varies with the different types of the inverted T-joint. The flow factor of H type joints tends to become larger with the brazing temperature, although the flow factor is generally small in this type joint. This seems to depend on the possibility that viscosity and the surface tension of the molten Cu-Zn alloy may decrease with increasing zinc content according to the elevation of brazing temperature. On the other hand, in the case of the V type joint specimen, the flow factor seems to decrease with the brazing temperature. The reason for this is that the molten Cu-Zn system alloy tends to flow onto the surface of horizontal bare base metal uncoated with the copper plating, and the increase in the amount of flowed alloy onto the surface may cause a decrease in fillet volume. In the W type joint, the test piece is covered all over with the Cu-Zn system alloy without a defect of partial flow; as the result, uniform and large fillets are obtained.

Figure 8 shows the outer views of brazed test pieces obtained by vapor phase brazing. Each seems to be by no means inferior to results obtained using ordinary methods of brazing. The Cu-Zn system alloy flows onto the surface of horizontal base metal, and can be observed in the V type test piece.

Figure 9 shows the microstructures of the joint interfaces brazed by the vapor phase brazing process. It is clear that the microstructures are affected by the brazing temperature. The microstructure of the fillet brazed at 1173 K (1652° F)

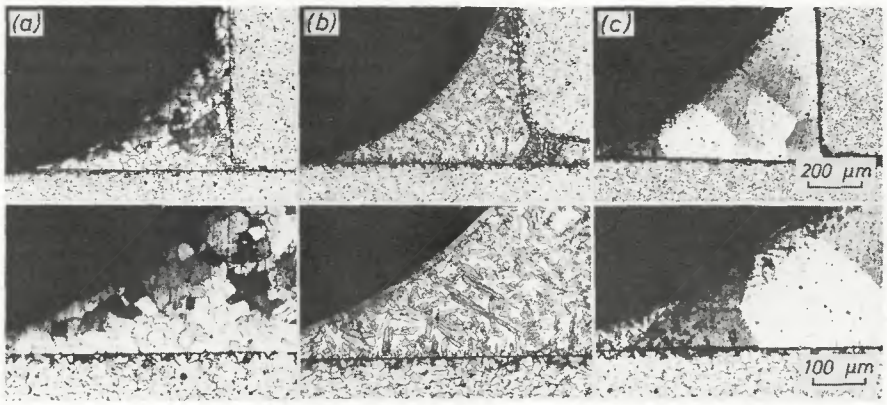

Fig. 9 — Microstructures of joint interfaces brazed using the vapor phase process at three different temperatures: (a) — 1173 K (1652° F); (b) — 1193 K (1688° F); (c) — 1213 K (1724° F)

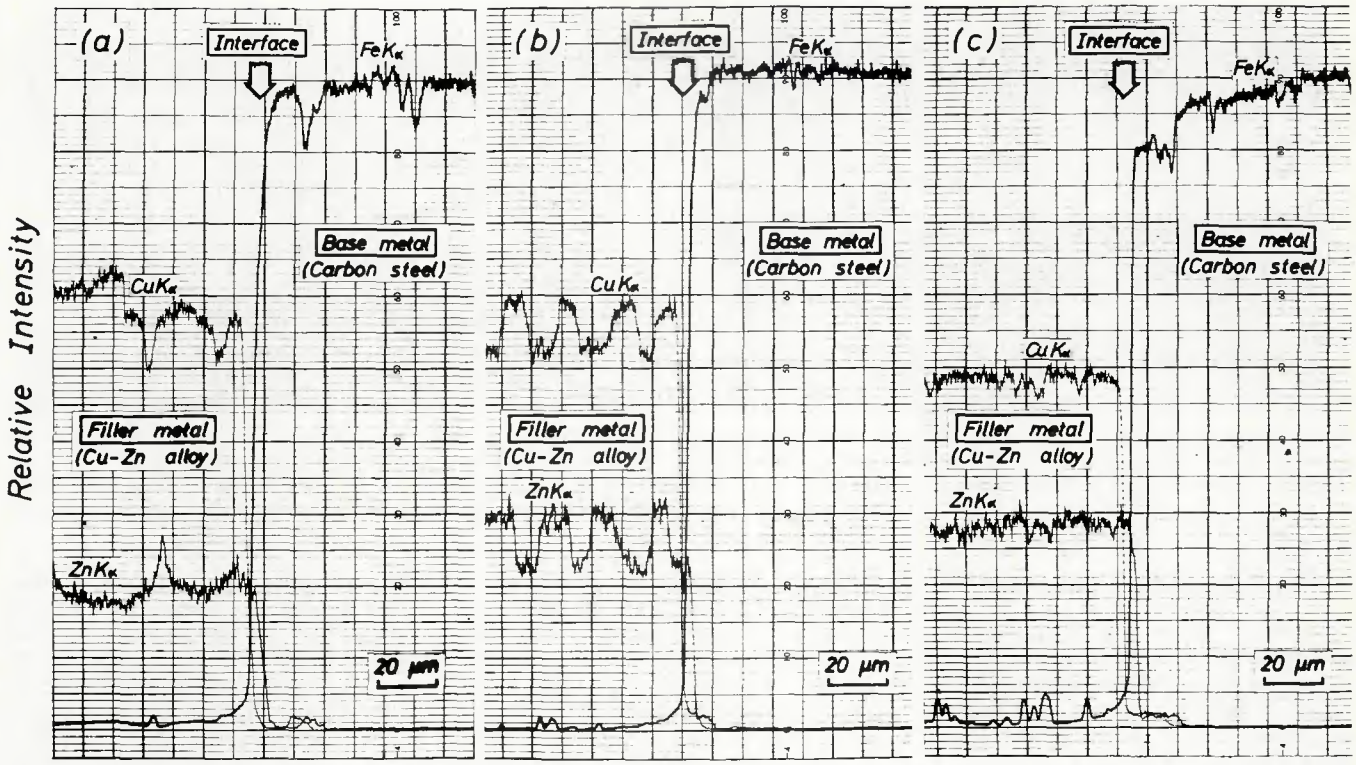

Fig. 10 — Patterns obtained by EPMA line anlysis for joint interfaces brazed at three different temperatures using the vapor phase process: (a) — 1173 K (1652° F); (b) — 1193 K (1688° F); (c) — 1213 K (1724° F)

seems to be composed of almost α phase, and the small fillet indicates the inferior flowability of the molten Cu-Zn system alloy as a "filler metal." The structure brazed at 1193 K (1688° F) seems to be the mixed phase of ($\alpha + \beta$), and the larger fillet indicates the superior flowability of a molten "filler metal." The structure brazed at 1213 K (1724°F) seems to be composed of the single β phase, and the intermetallic compounds of Fe-Zn system are observed at the interface.

Figure 10 shows the patterns obtained by EPMA for the brazed joint specimen. In the case of the joint brazed at 1173 K (1652° F), the zinc concentration is generally uniform all over the fillet, composed of the single α phase (Ref. 9). In the joint brazed at 1193 K (1688° F), the zinc concentration of the fillet is regularly alternated; judging from its concentration rate, it is considered that the fillet is composed of the mix phase of ($\alpha + \beta$). In the joint brazed at 1213 K (1724° F) the zinc concentration is almost constant; it is composed of the single β phase.

As mentioned previously, the results obtained by metallographic examination coincide with the results from the EPMA analysis for the Cu-Zn system alloy formed at the brazed joint.

Conclusion

The results obtained in the study described by this paper may be summarized as follows:

1. A newly developed vapor phase brazing process is suitable for carbon steel. Its brazing temperature is lower than the temperature normally encountered with other brazing methods, and the brazement is uniformly and completely coated by the gold-colored Cu-Zn system alloy.

2. The zinc contents of Cu-Zn system alloy obtained in the vapor phase brazing process were affected by the brazing temperature, and their metallurgical structures brazed at 1173, 1193 and 1213 K (1652, 1688 and 1724° F) were determined to be α phase, ($\alpha + \beta$) phase and β phase in the Cu-Zn equilibrium diagram, respectively.

3. The fillets formed at the brazed joints were faultless and by no means inferior to those obtained with ordinary brazing methods. The flow factor used to evaluate brazeability, was at the range of 0.7 to 0.9, and superior brazeability was observed.

References

1. Byrnes, Edward R. Jr. 1971. Vacuum fluxless brazing of aluminum. *Welding Journal* 50 (10): 712-719.

2. Schultze, W., and Schoer, W. 1973. Fluxless brazing of aluminum using protective gas. *Welding Journal* 52 (10): 644-651.

3. American Welding Society, 1976. *Brazing manual.* Miami.

4. Hansen, M. 1958. *Constitution of binary alloys.* New York: McGraw Hill.

5. Bredze, N., and Schwartzbart, H. 1959. Metallurgy of bonding in brazed joints. *Welding Journal* 38 (8): 305-s to 314-s.

6. Terrill, J. R. 1966. Diffusion of silicon in aluminum brazing sheet. *Welding Journal* 45 (5): 202-s to 209-s.

7. Kawakatsu, I., and Osawa, T. 1977. Furnace brazing of steel with brass filler. *Welding Journal* 56 (2): 56-s to 60-s.

8. Woods, R. A., and Robinson, I. B. 1974. Flow of aluminum dip brazing filler metals. *Welding Journal* 53 (10): 440-s to 445-s.

9. Toyota Central R&D laboratories, Inc. *Diagrams and tables for quantitative electron probe microanalysis.*