Studies on the properties of some copper-manganese alloys

A. K. LAHIRI, K. P. MUKHERJEE and T. BANERJEE

ESERVES of non-ferrous metals in India, except that of aluminium and manganese, are limited. Since resources of manganese metal are abundant in India, the possibilities of substituting manganese as a major alloying element in place of zinc and nickel in non-ferrous alloys are therefore important and need full applications. Manganese is one of the few elements which has a wide range of solubility in copper and the possibilities of utilising copper alloys containing manganese need careful consideration, as they have been shown to possess special properties, e.g., high strength, high damping property and high electrical resistivity. Work carried out at the U.S. Bureau of Mines¹⁻⁴ during the World War II showed the superior properties of different alloys made from electrolytic manganese compared to that of thermal manganese which invariably introduced impurities. Inspite of all these developments, the cost of electrolytic manganese and absence of sufficient data on the melting, working and various properties of these alloys have not yet encouraged any large scale production of these alloys.

The successful production of electrolytic manganese on pilot plant scale by the process developed and patented at the National Metallurgical Laboratory and the need for substituting imported metals, e.g. zinc, nickel, tin, etc., led to the decision to investigate in details the various binary and ternary alloys for their commercial exploitation in India. The results have been given in different publications^{5–8} and the present paper summarises the work with an idea to indicate the

possibilities of the field of substitution.

Phase diagram

Binary Cu-Mn system

Manganese occurs in four allotropic forms of which the tetragonal phase, stable between 1100-1136°C, is ductile. The low temperature phases, alpha and beta, having complex cubic structure, are extremely brittle. Copper is found to form a continuous solid solution

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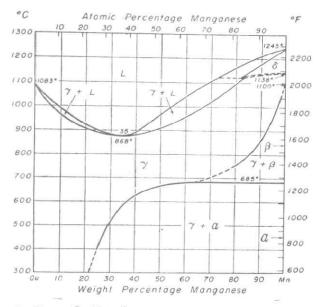
in gamma manganese at high temperatures (Fig. 1). The solubility decreases with decrease in temperature and is limited to about 20-25% Mn at room temperature. However, the rate of alpha or beta manganese is very sluggish and so as high as 80-90% manganese can be retained in solid solution at room temperature by rapid cooling. This metastable phase has a tetragonal structure with c/a ratio increasing with the increase in manganese content above 40%.

Ternary Cu-Mn-Zn system

Extensive survey of the copper-manganese-zinc system has been made for establishing solid solution boundaries and different phases present. According to Dean, the alpha field of copper-manganese-zinc system (Fig. 2) is bounded on two sides by copper-zinc and coppermanganese binary systems, and by the alpha-beta and alpha + X (zeta) fields on the other two sides. The most interesting part of this system is that it is possible to get a X single phase alpha alloy with as low as 50% copper and 10% zinc, rest being manganese. This is due to the fact that the alpha-beta boundary of the field remains approximately parallel to 60% copper up to 10% manganese and then curves towards the manganese corner with higher manganese contents. Increasing temperature moves the boundary towards the copper-manganese base line. X-phase appears only at low temperatures and is not detectable above 593°C. The alpha + X-boundary is almost perpendicular to copper-manganese base line; intersecting the base line at 27% and alpha + beta boundary at 10% manganese at 420°C. With increasing temperature this boundary is shifted to higher manganese content. The formation of the X-phase is sluggish and long time treatments are required for its appearance.

Melting and casting

Studies carried out indicated that copper alloys containing manganese are susceptible to gas porosity in casting, if proper care is not taken during melting. The porosity in the casting is mainly due to soluble gases and is due to reasons similar to those observed in melting of copper alloys containing tin, phosphorus

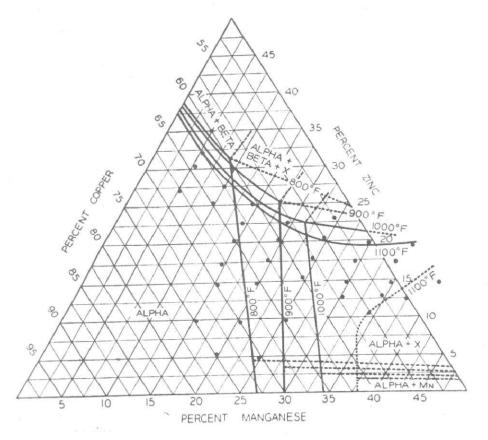


1 Binany Cu-Mn alloy system

and aluminium where hydrogen gas is the main culprit. To investigate the effect of different conditions, e.g., size and cross-section of the ingots and temperature of pouring on the degree of gas porosity studies were carried out in detail with an alloy of the composition Cu 75%, Mn 25%. The melts were made in oil-fired furnace without any slag cover and under oxidising flame.

Cross-section and height of the ingots were found to be of considerable importance in getting a sound ingot. With use of proper casting temperature increase in cross-section showed to decrease porosity indicating that a marked difference in solubility of gases in liquid and solid stages and a narrow range between liquidus and solidus play an important role in controlling porosity. Pouring temperature was found to be the most important controlling factor in this connection. Pouring temperatures of 1125-1025°C, the temperature decreasing with increasing manganese content, were found to be ideal to get sound ingot. In ternary alloys containing zinc, the degree of porosity was found to be considerably reduced. This was probably due to the scavenging action of zinc vapour rising up through the molten metal.

The effect of varying the amount of scrap material on the soundness of the ingots and the loss of alloying element were studied for binary Cu-Mn and ternary Cu-Mn-Zn-alloys in oil-fired furnace using various proportion of scrap and virgin metal. Loss of manganese and zinc, in alloys melted with 100% scrap and 50% scrap + 50%



Phase boundaries at 800°F (426°C), 900°F (482°C), 1000°F (532°C), 1100°F (503°C)

2 Specimens for 800°, 900° and 1000°F held at temperature for 48 hours and waterquenched. Specimens for 1100°F held at temperature for 4 hours and water-quenched

virgin metal, was high (12-15% manganese and 20% zinc) and the casting had a comparatively greater gas porosity. The extent of scrap used is also important from the point of view of carbon pick up by manganese when melting is carried out in graphite crucible or in arc furnace. In the case of melts with virgin metal, where manganese was added after copper was completely molten, carbon in the alloy analysed to be 0.03%. With 100% scrap this was raised to 0.12% as the molten alloy remained in contact with carbon for a much longer time. To avoid the pick up of carbon the extent of scrap addition should be kept at a minimum. Though no detailed studies were carried out to find out optimum conditions for melting scraps, it appears that scrap up to 25% of the charge can be used without much change in the melting technique.

Mechanical working

Hot rolling

Alloys could be hot-worked easily between 700°C and 900°C depending on the composition of the alloy, while the rolling temperature had to be kept below 850°C for alloys containing over 15% manganese. Some detailed studies were carried out on alloys containing 80% Cu-20% Mn. The factors studied were temperature of rolling and degree of reduction in each pass. Rolling temperature was varied between 900°C and 650°C. Alloys heated above 850°C were found to hot-crack during rolling. Rolling temperature between 800°C and 850°C was found to give better rolled surface. Ingots rolled at or below 700°C gave excessive cracking and great load on the rolling mill. The condition of $1\frac{1}{2}$ "x $1\frac{1}{2}$ " billets rolled from 4" dia. ingots at different temperatures is shown in Fig. 3.

Cold rolling

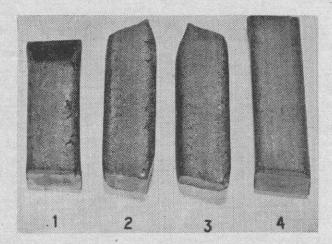
The copper-manganese alloys (even those containing 30-40% Mn) were found to be easily cold-rolled to the extent of 75% reduction in thickness. Ternary alloys containing nickel were quite tough but the same containing zinc were comparatively more amenable to both hot and cold working.

Studies on properties of alloys

Material preparation

For the present studies, eight groups of alloys O,A, B,F,C,E,D and N (alloys containing nickel) were selected having nominal copper content of 95, 90, 80, 75, 70, 65, 60 and 70 per cent respectively. Except the group No. N, which contained nickel, the others were binary copper-manganese and ternary copper-manganese-zinc alloys. The manganese, zinc and nickel varied between 5-40%, 5-25% and 0-8% respectively. The analysis of alloys studied is given in Tables I and II. In the group containing nickel, one composition of quarternary copper-manganese-zinc-nickel alloy was also included.

All the alloys studied, except D2 and D4, fell within



Rolled at 700°C;
 Rolled at 750°C;
 Rolled at 800°C;
 Rolled at 825°C.

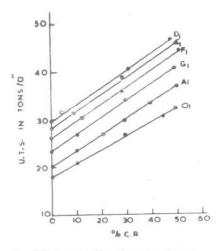
3 Cracking of hot rolled alloys

TABLE I

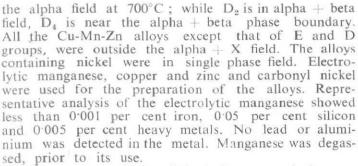
Alloy No.	% Comp				
	Cu	Mn	Zn	Ni	Remarks
	v				
O_1	95.04	4.94	-	_	
A_1	89 33	10.64			
\mathbf{B}_1	78-53	21.38	_		Binary
\mathbf{B}_2	81.00	9.63	9.17		Ternary
F_1	75.40	25.50		_	
C_1	70.81	29.15		_	Binary
C_2	72.85	20.71	6.14	_	Ternary
C_3	69.65	16.65	13.80	_	,,
C ₄	70.42	9-73	19.56		,,
E ₁	64-94	8.50	26.45		,,
E ₂	64.15	25.46	10.33	_	,,
D ₁	56.69	40-21	_		Binary
D ₂	55.74	27.96	15.66	_	Ternary
D_3	59.49	23.63	17:30	_	,,
D_4	58.68	19:49	20.84		.,,

TABLE II

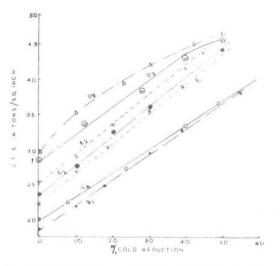
Allow	% Compo	osition			
Alloy No.	Cu	Mn	Ni	Zn	
N ₁	69.52	24.8	4.94		
N ₂	69.52	22.16	7.70		
N_3	59-1	25.13	4.9	11.0	



4 Effect of cold rolling on U.T.S.

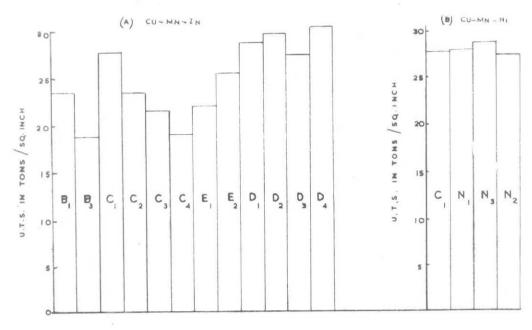


Alloys were made in oil-fired furnace and the procedure consisted of melting of copper, addition of



6 Effect of rolling on U.T.S. of Cu-Mn-Zn alloys

manganese and deoxidation with 0.2% aluminium prior to casting. In case of alloys containing zinc, the latter was added after deoxidation. No slag cover was used for melts. Additional amounts of 5% and 10% of the respective weight of manganese and zinc were added to compensate for melting losses. The alloys were cast in slabs of 8" × 10" × 1" size through a tundish. A close control of teeming temperature was kept to avoid gas porosity. The slabs were hot-rolled at 800°-725°C depending on composition and then cold-rolled with intermediate annealing to produce 0.06" thick sheets, which were used as the starting material for different studies. Prior to cold-rolling, the hot-rolled sheets were pickled in solution containing 15%



5 U.T.S. of alloy annealed at 700°C

H₂SO₄ 15% HNO₃ and 70% water at 40°-45°C.

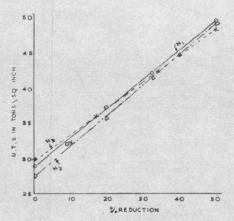
Mechanical properties

Tensile strength, elongation and hardness were determined for all the alloys in annealed and cold-worked conditions. Standard tensile specimens with a 1"-gauge length were used. The properties after cold-working were determined on sheets (annealed at 700°C for 1 hour) cold-rolled to 10, 20, 30, 40 and 50 per cent in thickness. The properties as affected by annealing at different temperatures were determined on 50% cold-rolled material heated for 2 hours at temperatures ranging from 300°-750°C followed by quenching. At least, two specimens, produced under identically the same conditions, were tested. Some of the properties of different alloys are given in Table III and plotted in Figs. 4-7.

Physical properties of alloys annealed at different temperatures

The strength of annealed Cu-Mn alloys increases with increase in manganese content, first rapidly and then slowly between 20 and 40% Mn (Fig. 5). The hardness values of the alloys ran largely parallel to their strength. However, no corresponding marked change in percentage elongation was observed. While the alloy O_1 showed 52-54% elongation, alloys B_1 to D_1 showed on the average 47.5% elongation when quenched from 750° C.

In case of ternary systems, the property differences between different groups of alloys were noted in the annealed materials. With decreasing copper and increasing manganese content higher tensile strength and hardness were obtained. For groups B, C and E (Fig. 5) increasing the zinc had an effect in decreasing the tensile strength. The effect on hardness was similar but not so marked as on the tensile strength. Percentage



7 Effect of cold rolling on U.T.S. of Cu-Mn-Ni alloy

of elongation was, however, not affected to the same extent and difference between binary and ternary alloys was found to be quite small.

In alloys of group D containing 60% Cu annealing at 700°C showed either slight decrease or increase in tensile strength with increasing zinc content depending on whether the alloys were single phase $(D_1 \text{ and } D_3)$ or double phase $(D_2 \text{ and } D_4)$.

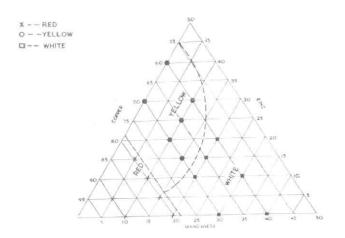
In alloys of copper-manganese-nickel group, the addition of nickel increased tensile strength and hardness to a small extent while the percentage of elongation was not much affected. Replacement of manganese partly by zinc had the effect of decreasing the strength and hardness.

Effect of cold working

All the alloys showed expected increase in hardness and strength, and decrease in percentage elongation with

TABLE III Change in mechanical properties on annealing

			Mechanical properties after annealing at (°C)					
	Cold Rolled		300		400		700	
Alloy No.	Hardness in V.P.N.	Percentage Elongation	Hardness in V.P.N.	Perncentage Elongation	Hardness in V.P.N.	Percentage Elongation	Hardness in V.P.N.	Percentage Elongation
B ₃ C ₂ C ₈ C ₄ E ₁ E ₂ D ₂ D ₃ D ₄	188 218 202 210 202 211 248 223 236·8	6·0 3·0 6·0 6·0 6·6 3·0 2·0 3·0 2·0	192 222 214 210 207 — 249 228 235	8·0 6·5 8·5 8·5 10·0 6·5 3·5 6·5 3·5	119·5 131·5 127·5 121 125 132·4	33·5 27·5 33·5 37·5 37·5 27·5 23·5 27·5 19·0	75 86 78 72 76 88 106 99	52:5 47:5 50:6 52:5 55:0 47:5 33:5 44:5 37:5
N ₁ N ₂ N ₃	228 218 230·5	6·0 6·0	228 220 231	8·5 10·0 8·5	131 129 135	33·5 37·5 27·5	87 82 92	50·0 52 47·5



8 Regions showing red, yellow and white colour of copper, manganese and zinc alloy system

increasing degree of cold-working. Tensile strength of alloys after different degrees of cold-rolling (plotted in Figs. 5, 6 and 7 for some of the alloys) showed linear increase up to 50% reduction. The slope of the curve was found to vary with composition. In any particular group of alloys, the rate of work-hardening was found to be more for these containing higher manganese. Unlike other groups, alloys D₂ and D₄, which contained both alpha and beta phases, did not show linear rate of work-hardening.

Deep drawing test

Copper and many of its commonly used alloys possess

good forming properties. Fabrication stage in sheet alloys involves deep drawing and stretch forming invarying degrees. Relative deep drawing characteristics of the present alloys were therefore determined by Swift's 6½" cupping press using flat punch and compared with that of brass.

The deep drawing property was defined by a term L. R. D. and the maximum pressure for drawing, where:

L. R. D. = Maximum dia. of the blank drawn successfully Dia. of the plunger

For the present studies, sheets were cold-rolled to a thickness of 0.045-0.046", to give a smooth finish cleaned in bright pickling solution and then used for cupping test. The circular plates were clamped to the die-face by applying a pressure of 80 lbs/sq.in. to the blankholder plate. Plunger of 2" dia. was used. A successful draw was considered to be the one which was completely drawn, without wrinkles. The result of the deep drawing test is given in Table IV, which shows that manganese lowers the deep drawing response of copper more than zinc. Addition of zinc had a marked effect in improving the deep drawing property, though nickel did not have similar effect. The maximum load (200-475 lb/sq.in.) varied with composition and was found to be more for alloys containing lower zinc and higher nickel and manganese. The alloy E₁, which showed very good deep drawing property, had slightly greater drawing load than brass. The poor drawability of D group of alloys was probably due to their high manganese content and to some extent due to presence of beta phase in the alloy. Unlike zinc, nickel had no such marked effect on deep drawing properties. The overall deep drawing property of alloys containing manganese up to 20% was quite adequate and so these alloys were amenable to forming operations.

TABLE IV Swift's Cupping test results (a) For Cu-Mn-Zn alloys (annealed at 700°C)

hickness n inches	3.8′′	3.9"			Blank size (inches)							
		33	4.0′′	4.1′′	4.2′′	4.3"	4.4′′	4.5′′	4.6"	load in lb/in²	dia, in inches	L.R.D.
0.047		_			S	S	S	F		300	4.4	2.2
0.046	-	_		S	S	S		-	(0.000)	400	4.3	2.15
0.046		-	-	S	S	S	F	-		400	4.3	2.15
0.047	-			-		S	S	F	-	350	4.4	2.20
0.047		-	-		S	S	S	S	F			2.25
0.046	S	S	S			-			-			2.0
0.046	S	S	S	S	S	F	-					2.1
0.047	S	S	F	F		-	-	7000	2 -1	450	3.9	1.95
			(b) F	or Cu-N	In-Ni allo	y (anneal	ed at 75	0°C)				
0.047	_		_	S	S	S	F		_	475	4.3	2.15
0.046	-	-	200	S	S	S	F	-	-	475	4.3	2.15
0.046			_	S	S	S	F			425	4.3	2.12
000000	046 047 047 047 046 046 046 047	-046 — -047 — -046 S S -047 S S -047 — -046	1046 — — — — — — — — — — — — — — — — — — —	1046 — — — — — — — — — — — — — — — — — — —	1046 — — — — — — — — — — — — — — — — — — —	1046 — — — S S S S S S S S S S S S S S S S	1046 — — — S S S S 1046 — — — S S S S 1047 — — — S S S 1047 — — — S S 1047 — — — S S 1046 S S S S F — — — 1046 S S S S S S S S F 1047 S S F F — — — 1047 — — — S S S 1047 — — — S S S 1047 — — S S S S	1046 — — — S S S F F 1046 — — — S S S S F F 1047 — — — — S S S S S S S S S S S S S S S	1046 -	1046 - S S S F	1046 -	1046 -

S-Success; F-Failure.

Colour

Increasing manganese brought about a definite colour change in the alloys. The variation in colour with composition is shown in Fig. 8. With increase in manganese the binary alloy became progressively lighter in colour and above 20% manganese it became completely white. In ternary system addition of zinc had an effect in changing the white binary copper-manganese alloys to yellow; the amount of zinc required increased with decrease in the copper content. Unlike the binary alloys, which were of only two colours, i.e., white and red, the ternary alloys showed all the three colours (red, yellow and white).

All the nickel containing alloys investigated were white in colour, because of the fact that manganese content in all of them was above 20%.

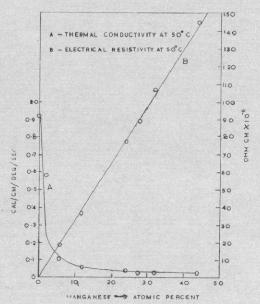
Corrosion studies

Salt spray test

Weight loss data of samples after exposure in salt spray chamber for 120 hours (data of binary alloys are for 100 hours) at $35\pm1^{\circ}$ C, with a 5% sodium chloride solution are given in Table V. The tests were run intermittently consisting of 8 hours spraying time. For rest of the period, the samples were retained inside the chamber under high humidity condition. The weight loss showed that addition of manganese increases corrosion rate of copper specially when present above 10%. For ternary alloys addition of zinc and nickel considerably increased the corrosion resistance of binary copper-manganese alloys; the effect of nickel being greater than that of zinc.

TABLE V Results of salt spray test

Solution period	5% NaCl 120 hours (daily 8 h spraying)			
Alloy group	Alloy no.	Weight loss in gm/dm ²		
Cu-Mn	$O_1 \\ A_1 \\ F_1 \\ B_1 \\ C_1 \\ D_1$	0·094 0·105 0·145 0·127 0·166 0·192		
Cu-Mn-Zn	$\begin{array}{c} B_3 \\ C_3 \\ C_2 \\ C_4 \\ E_1 \\ E_2 \\ D_2 \\ D_3 \\ D_4 \end{array}$	0·120 0·088 0·095 0·087 0·090 0·102 0·133 0·112 0·105		
Cu-Mn-Ni	N ₁ N ₂ N ₃	0·09 0·08 0·065		
Brass (30% Zn) Cupro-nickel (25% Ni)		0·105 0·022		



9 Thermal conductivity and electrical resistivity of Cu-Mn alloys

Stress corrosion cracking studies

Stress corrosion cracking susceptibility of binary and ternary copper-manganese-zinc alloys has been studied by Lahiri and Banerjee.^{7,8} The cracking susceptibility of cold-rolled alloys and of the same annealed at 300°C and 400°C has been compared in the present paper.

Strip samples of size 12.7 mm × 76.2 mm were given a constant strain by defecting the strip at the centre over a 5.33 mm dia. glass rod and fixing the ends to plastic specimen holder by means of mild steel bolts. The samples were exposed, immediately after stressing, to ammonia atmosphere inside a desiccator. Specimens were taken out at different time intervals to observe the appearance of crack under a low power microscope (×20).

Cracking time and nature of cracking for different binary and ternary copper alloys have been given in Table VI. For comparison, values for alpha Cu-35 Zn brass have also been included in the above table. The results indicated that compared to brass, alloys containing manganese have higher resistance to cracking. Addition of zinc progressively decreased the cracking time of copper-manganese alloys. Alloys containing 60% copper (Group D) showed very high resistance to cracking and except D₃, which contained 21% zinc, none of the other alloys failed.

Electrical and thermal conductivity

Manganese has been known to increase the electrical resistivity of copper long before the solid solution nature of these alloys were established. The most commonly used resistant coils of constant resistivity is manganese, which is a Cu-Mn-Ni alloy. Gupta and Banerjee⁹ have studied in detail the electrical and thermal

TABLE VI

2.44		Cracking time	in hours for alloys ar		
Alloy No.	Nominal Composition	Cold-rolled	300°C	400°C	Nature of cracking
3,	80 Cu-20 Mn 70 Cu-30 Mn	32 720 (N.C.)	28 720 (N.C.)	60 720 (N.C.)	Transgranular
В	80 Cu-20 Zn	3.5	ì·75	2:5	Transgranular in cold-rolled con- dition. Intergranular when annealed.
В	70Cu-30 Zn	3.5	1.75	2.5	
2	85Cu-7.5Mn-7.5Zn	42	24	28	Intergranular
3	80Cu-10Mn-10Zn	16	15	20	do
2	70Cu-20Mn-10Zn	56	1 <u></u>	40	Transgranular
4	70Cu-10Mn-20Zn	50	14	34	Trans, when cold-rolled and inter, when annealed.
2	65Cu-25Mn-10Zn		No Cracking		Care Service Committee Com
1	65Cu 10Mn-25Zn	30	6	4	Trans, when cold-rolled and inter, when annealed.
)3	60Cu-25Mn-15Zn 60Cu-20Mn-20Zn	48	No Cracking No Cracking		Transgranular —

N. C .= No Cracking

conductivity of Cu-Mn alloys up to 40% manganese. The results are plotted in Fig. 9, which shows that electrical resistivity increased linearly with atomic per cent manganese in the range studied. In the temperature range of $0-100^{\circ}\text{C}$, the temperature coefficient of electrical resistivity decreased sharply from 43×10^{-4} to 1.35×10^{-4} with change in manganese content from 0 to 5%. With further addition of manganese the temperature coefficient first decreased and then again increased. The minimum in the temperature coefficient was between 10-20% Mn, the value being of the order of 0.03×10^{-4} .

The thermal conductivity of copper decreased appreciably even when Mn is present in small amounts in copper-manganese alloys. 10% manganese reduced the thermal conductivity to 6.5% of that of copper; with further increase in manganese the conductivity went on decreasing, and at 40% manganese the thermal conductivity was reduced to 3.3% of that of pure copper.

Conclusion

The studies have presented interesting data on the physical, mechanical and corrosion resistance properties of copper alloys containing manganese indicating the possibilities of their substitution in many cases where conventional brass is used. On the basis of these studies it can be said that:

- 1. In binary alloys, addition of manganese progressively increases the hardness and tensile strength of the alloy, at first rapidly and then slowly.
- The alloys possess good hot and cold working properties.
- Compared to binary copper-manganese alloys, ternary alloys containing zinc possess lower strength and hardness. However, these are stronger

than copper-zinc alloys. Alloys containing nickel are, however, stronger than those containing zinc and addition of nickel progressively increases the hardness and tensile strength of Cu-Mn alloy. The percentage elongation of the single phase ternary alloys is dependent on zinc content, it increasing with increase in zinc content.

- All the alloys possess adequate deep drawing properties suitable for forming operations.
- The corrosion resistance of copper-manganese alloys decreases with increase in the manganese content which can be considerably improved by the addition of zinc and nickel.
- The alloys containing manganese are in general more resistant to stress corrosion cracking than copper-zinc alloys.

The single phase alloys show high strength combined with good ductility and deep drawing properties in consideration of which these can be substituted for conventional brasses, such as bolts and tie-rods, shafting, rivets, valves, automobile lamp bodies, lamp bases, domestic and industrial plumbing, etc. The possibilities of using alloy 65 Cu-25 Zn-10 Mn, can be examined for the cartridge cases as also suggested by Dean. These can be used to substitute manganese bronze where the corrosive conditions are not very severe. There are many other possible uses of brass, e.g., pipes for oil refineries and utilities, shaft for marine uses, water meters, centrifuges in sugar industry, etc., where they can be substituted by copper alloys containing Mn but to ascertain this a more detailed study on the corrosion resistance properties under different conditions is required, which is under progress at the National Metallurgical Labora-

Acknowledgements

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Discussions

Mr Uday Chatterjee (NML): Stress corrosion properties: A series of alloys extending from 3-22% Mn in the wire form have been tested in copper ammonium sulphate solution. A distinct minimum has been shown in the cracking time vs %Mn plot for an alloy of nearly 15% Mn (Solution pH=6.9).

From the experiments carried out it has been found

that the solution pH has a marked effect on the cracking susceptibility of these alloys. There are also indications that this minimum may be shifted to a higher Mn side subject to solution pH.

Further work in this line is now in progress at the National Metallurgical Laboratory.