Assessment of structure and selected mechanical properties of braze welded joints of copper-lined steel tubes

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Abstract

In the paper, results of a research on braze welded joints of copper-lined 10CrMo9-10 (10H2M) steel tubes made by CMT arc braze welding are presented. As a filler metal, silicon bronze was applied. Results of metallographic examinations with the use of light microscopy, electron scanning microscopy and X-ray microanalysis are presented. Mechanical properties of the joints were assessed on the grounds of static tensile test, technological bending test, as well as hardness measurements. Quality of the obtained joints was good. No cracks were observed in the brazed joints and the obtained mechanical properties were satisfactory.

Keywords 10CrMo9-10 steel · Cu-ETP copper · CuSi3 filler metal · CMT braze welding · Braze weld · Reaction zone · Inclusions in braze weld · Metallographic examinations · Mechanical properties of joints

1 Introduction

Joining dissimilar materials is related to various difficulties that can basically affect quality of the obtained joints. In such a case, the main difficulties are of metallurgical nature and occur especially in the welding processes, where components of base materials and the filler metal are alloyed together, i.e. in the so-called fusion welding [1]. This may cause the formation of hard reaction zones [2] or hot cracks [3]. This problem occurs also during bonding copper with steel [4]. Joints of these materials are, nowadays, widely used in various industries, among others in cooling appliances, heating systems, boilers, household appliances, structures built for chemical, shipbuilding and metallurgical industries [5], as well as in windings of high-power generators [6].

In the case of joining copper with steel, the main problem results from limited solubility of copper in steel [7].

Maximum solubility of copper at high temperature is 6.5 wt% in δ iron, 8 wt% in γ iron and 1.4 wt% in α iron [8]. At ambient temperature, at phase equilibrium state (Fig. 1), solubility of copper in iron is limited to 0.3 wt% only [7].

This restricts application of traditional methods of arc welding, commonly used in industry, i.e. the methods MIG and TIG [9]. An alternative for them can be low-energy versions of GMA (Gas Metal Arc) welding: ColdArc, STT (Surface Tension Transfer), CMT (Cold Metal Transfer), etc. [10], with synergic lines dedicated for the brazing process [11]. The low-energy arc joining processes are dedicated for materials sensitive to heat [12] and joining thin sheets [13]. Application of braze welding significantly reduces the possibility of hot cracking, since, in this process, partial melting of the joined materials and mixing their components with the filler metal do not occur or are considerably reduced and the joint is obtained mainly by wetting and diffusion, like at brazing (less often at soft soldering) [10, 11]. The way of preparing the materials for braze welding is identical to that applied in the welding process, i.e. with a gap or chamfered edges, with no capillary gap, characteristic for brazing [11].

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Fig. 1 Phase equilibrium diagram Cu–Fe [7]

Fig. 2 10CrMo9-10 steel tube (1) with internal Cu-ETP lining (2)

2 Materials for braze welding

The base material to be examined was a steel tube grade 10CrMo9-10 (1.7380) with outside diameter 101.6 mm and wall thickness 15.5 mm, with internal copper lining 2-mm thick (Fig. 2). The tube is designed for application

in the chemical industry at 550 °C and under pressure of 40 bar. The internal copper lining is mechanically forced to the steel tube and protects steel against corrosive action of aggressive working medium.

According to EN 10216-2:2014-02, the 10CrMo9-10 steel belongs to the group of steels designed for operation at elevated temperatures. It is mainly applied in manufacture of steam boilers, steam turbines, steam pipelines and pressure appliances. The Cu-ETP copper, of which the internal lining was made, is classified as “electrolytic tough pitch copper” with controlled low content of oxygen (copper oxide). It is characterised by very good thermal and electrical conductivity, good corrosion resistance in various environments and resistance to most aggressive media except those containing ammonia. It is widely used in electrical industry, in sanitary systems and in automotive industry.

Selection of a filler metal for braze welding dissimilar materials is a big problem. Generally, its melting point corresponds to that of the lower-melting base material [10, 11]. If the difference between melting points of the filler metal and the base material is small, it is not always possible to avoid partial melting of the edge of the lower-melting material [14]. For brazing the 10CrMo9-10 steel tube with copper lining, the silicon bronze CuSi3 in the form of dia. 1.0-mm wire was selected.

Chemical compositions of the medium-alloyed steel 10CrMo9-10 acc. to EN 10216-2:2014-02, Cu-ETP copper acc. to EN 13599:2014-04 and the filler metal CuSi3 acc. to EN ISO 24373:2009 are presented in Table 1.

Depending on cooling rate, different microstructures of the 10CrMo9-10 steel can be obtained: bainitic after air cooling (normalising), ferritic-bainitic after slow furnace cooling (annealing) and martensitic after quenching in oil. Bainite obtained in this steel is globular (carbide free) and is subjected to tempering, like martensite.

Microstructure of the examined steel 10CrMo9-10 is ferritic-bainitic with diversified ferrite grain size and with slight banding (Fig. 3).

3 Fabrication of braze welded joints

Preparation of the tube for braze welding by “V” chamfering its edges is shown in Fig. 4.

Table 1 Chemical compositions of joined materials and filler metal

Material	Chemical composition (wt%)														
	C	Mo	Si	Mn	Cr	Ni	P	S	Sn	Zn	B	O	Pb	Cu	Fe
10CrMo9-10	0.08 ÷ 0.15	0.9 ÷ 1.1	0.15 ÷ 0.50	0.4 ÷ 0.6	2.0 ÷ 2.5	—	< 0.03	< 0.03	—	—	—	—	—	< 0.25	Rem
Cu-ETP	—	—	—	—	—	< 0.3	—	—	—	—	0.0005	0.04	0.005	Rem	—
CuSi3	—	—	< 3.0	< 1.0	—	—	—	—	< 0.1	< 0.1	—	—	—	0.07	Rem

Fig. 3 Ferritic-bainitic microstructure of 10CrMo9-10 steel. Etched with Nital

Fig. 4 Preparation of copper-lined 10CrMo9-10 tube for braze welding

With regard to small quantity of the material (steel tube with copper lining) and results of the first braze welding trials, the tubes were not joined on their entire circumference, but were cut lengthwise into 4 equal pieces—quarters, each 90-mm long. Before braze welding, thin copper plates—run-on and run-off ones—were TIG-welded to the tube quarters (Fig. 5). The TIG welds were made only between the inner copper pipe and the copper plates. This was done with the introduction of the minimum welding linear energy and was necessary for the experiment, in which only a quarter of the circumference of the pipe was braze welding. Under normal braze welding conditions, this type of pipeline is not necessary.

The braze welding process was carried out on a robotized station, using the low-energy method CMT, under shielding of pure argon 4.5 (99.995 vol% Ar).

The station was equipped with a synergic power source Fronius TransPuls Synergic 3200 with a digital control of welding current, a system responsible for wire withdrawal at the moment of a short circuit to minimise spattering and a digitally controlled wire feeder Fronius VR 7000-CMT

Fig. 5 Preparation of specimens for braze welding: 1—10CrMo9-10 steel tubes, 2—Cu lining, 3—run-on plate, 4—run-off plate, 5—fixing holders

4R/G/W/F++. Working movements during the process were executed by the robot Kawasaki series BA with a welding clamp Robacta Drive CMT PAPW fitted on its arm end, with an integrated, numerically controlled, asynchronous AC motor permitting quick withdrawal of wire to the special buffer [15].

The braze welding parameters were selected on copper plates of 2.0-mm thick. It was started from the parameters implemented in the synergic line for the filler metal CuSi3 and copper as the base material with a preset thickness. Next, these parameters were corrected by changing the braze welding speed. The preliminary trials demonstrated that the process requires a very precise selection of process parameters, whose change—even within a very narrow range—decreases the process stability and results in numerous imperfections in the joints. Even small changes of parameters resulted in, e.g. spattering due to lower process stability, lack of wetting and bonding or excessive partial melting (melt-through) of copper edges. Final parameters of CMT braze welding, for which two joints (Nos. 1 and 2) were made, are shown in Table 2. The welding groove was completely filled by application of 15 seams. After execution of trial joints, the run-on and run-off plates were removed and then specimens for metallographic macro- and microscopic examinations and for mechanical testing were cut out.

4 Radiographic examinations

Radiographic testing (RT) was carried out according to the standards EN ISO 17636-1:2013-06 and EN 12799:2003/A1:2005. The radiographs executed on $\frac{1}{4}$ circumference of tubular joints did not show any internal imperfections in the braze welded joints. An exemplary radiograph is shown in Fig. 6.

Table 2 Parameters of braze welding copper-lined tube of 10CrMo9-10 steel

Item	Parameter	Value
1	Current intensity, I	178 A
2	Arc voltage, U	18 V
3	Wire feeding rate, Vd	10.8 m/min
4	Braze welding speed, V_s	60 cm/min
5	Wire extension, L_w	15 mm
6	Argon flow rate, Q	16 m ³ /min

Fig. 6 Exemplary radiograph (negative) of a braze welded joint of copper-lined 10CrMo9-10 steel tube

5 Macroscopic examinations

Polished sections for macro- and microscopic examinations were prepared in a standard way. The specimens were ground on abrasive papers and next polished using diamond pastes. Macroscopic examinations were performed with an unaided eye and with use of a stereoscopic microscope MST-800 Nikon.

It can be seen in Fig. 7 that the joint was made in a correct way, with no visible macroscopic imperfections. A missing connection between the steel tube (1) and the copper lining (2) on the whole length of the tube (which results from mechanical assembly of steel tube and lining by forcing in) is visible. However, the connection exists in the braze welded place. A small partial melting can be seen at the weld root and at the weld face (A, B). Braze welded joints are assessed like brazed joints; so according to EN ISO 18279:2008, this is an imperfection 7UAAC of the group VI “Miscellaneous imperfections”, i.e. the excessive reaction of the filler metal with the base material. In dissimilar joints, especially of steel and copper, it can be important, which of the materials undergoes partial melting. Chemical composition of the filler metal is similar to that of the copper lining; so it is more favourable that copper is partially melted. In the case when partial melting

Fig. 7 Macrostructure of braze welded joint No. 1: 10CrMo9-10 steel tube (1), copper lining (2), braze weld (3), missing connection between steel and copper lining (4), partial melting of steel (A, B). Etched chemically with Nital and with a solution of chromic and nitric acids

of steel occurs, hot cracks can emerge as a result of copper diffusion from the filler metal to the steel along grain boundaries, which was presented in [16]. Formation of hot cracks could be also favoured by quick dissipation of heat from the joint area by the copper lining, which increases tensile stresses in the steel and facilitates diffusion of copper from the deposited metal along grain boundaries. In the considered case, no macroscopic hot cracks were found in steel.

6 Microscopic examinations and EDS microanalysis

Microscopic examinations were carried out using a light microscope Olympus C40M with image recording by a digital CCD camera.

For the examinations, two braze welded joints Nos. 1 and 2 were chosen. Observations were performed in various areas of the weld and the joined materials. Since they are braze welded joints created by diffusive mechanisms, their functionality and mechanical properties depend, to a high degree, on quality of the connection of the boundary between the braze weld and the 10CrMo9-10 steel. In the central part of the joint No. 1, both on the left and on the right of the weld, a reaction zone with variable width is visible, created as a result of a dissolution of the steel into the braze weld as well as partial melting of steel (Fig. 8).

The reaction zone on the left side of the weld (Fig. 8a) is ca. 4 times wider ($10 \div 15 \mu\text{m}$) than on the opposite side (Fig. 8b). It was caused by position of the welding clamp (from that the electrode wire was advanced) in relation to the edge of the welding groove and/or the sequence of applying individual seams. The first seam was applied on the right side, so—when applying the seam on the left side—the base

Fig. 8 Microstructure of brazed joint No. 1: 10CrMo9-10 steel tube (1), braze weld (2), reaction zone (3), inclusions in the weld (4). Left side of the weld (a); right side of the weld (b). Etched chemically with Nital and with 10% solution of ammonium persulphate

Fig. 9 Microstructure of brazed joint (a) with inclusions from Fig. 8a (material contrast) and characteristic X-ray spectra for the inclusion (b) and the matrix (c). Primary beam energy $E_0 = 20$ keV

material was heated to a higher temperature, which is conducive for the diffusion mechanisms responsible for creation of the reaction zone. A slight partial melting of steel could also take place. Tiny inclusions are present in the braze weld, whose larger amount can be also observed on the left side of the joint—with wider reaction zone.

SEM examinations and EDS microanalysis were carried out using a scanning electron microscope JEOL JSM6610 equipped with an EDS spectrometer. Local qualitative and quantitative analyses were also performed using a

standardless programme ZAF, and element distribution maps were recorded.

Inclusions in the braze weld were identified as an intermetallic phase, rich in Fe and Si (Fig. 9 and Table 3). Tiny inclusions created by precipitating from a liquid filler material in which Fe coming from partially melted steel has been dissolved.

From the Si side of the Fe–Si system (Fig. 10a) [17], 3 silicon solutions in iron occur: disordered solution α (marked as αFe in the diagram) up to 10 at.% Si at ambient

Table 3 Results of local quantitative analysis for the braze weld from Fig. 9a

Point no	Location	%	Element			
			Cu	Fe	Si	Mn
1	Inclusion	wt%	5.2	77.6	14.4	2.9
		at. %	4.00	68.2	25.2	2.6
2	Matrix	wt%	95.5	2.7	2.7	0.4
		at. %	93.5	3.0	3.0	0.4

Fig. 10 A fragment of the phase equilibrium diagram Fe–Si [17] (a) and isothermal section through the diagram for the system Fe–Cu–Si [19] (b)

temperature, ordered solution α_2 within 10 and 11% Si and ordered solution α_1 within 11 and 25% Si. The ordered solutions are treated as intermetallic phases. The crystal lattice network B2 is attributed to the phase α_2 and the lattice D0₃ is attributed to the phase α_1 [18]. In the braze weld, the three-component system Fe–Si–Cu is present. Silicon only shows good solubility in iron, but solubilities of Si in Cu, Cu in Fe and Fe in Cu are limited and, in the Fe–Si–Cu system, two- or three-phase mixtures are mostly present (Fig. 10b) [19]. Figure 10b shows an isothermal cross section of the phase equilibrium diagram Fe–Si–Cu at 750 °C, but it can be supposed that differences of phase constitution at ambient temperature will not be large. In the braze weld and in the reaction zone, mixtures of various solutions of Si in Fe

(marked as α Fe in Fig. 10b), as well as solutions of Fe and Si in Cu (marked as Cu in Fig. 10b) can be expected. Table 3 shows results of EDS quantitative microanalysis of the braze weld in its central part. The structure is composed of the matrix being a solution of Fe and Si in Cu and particles of the phase α_1 . Analysis of the inclusions shows also the presence of Cu, which can indicate that the particles are not one-phase, but, in addition to the α_1 phase, they contain very tiny particles of a Cu-based solution. The braze weld contains also manganese coming from the filler metal and from steel. This element is mostly concentrated in the excluded particles. Manganese should not significantly affect properties of the braze weld.

Fig. 11 Microstructure of braze welded joint No. 2: 10CrMo9-10 steel tube (1), braze weld (2), reaction zone (3), inclusions in the braze weld (4). Left side of the weld in the central part (a), right side of the weld in the central part (b), left side of the weld at the root (c), right side of the weld at the root (d). Etched chemically with Nital and with 10% solution of ammonium persulphate

In the central part of the joint No. 2, on the boundary between steel and the braze weld, width of the reaction zone is comparable on both sides (Fig. 11a, b). In the braze weld itself, much smaller amount of the Fe-containing phase can be observed, which evidences much lower degree of its partial melting in this weld zone. Further in the direction of the weld root (Fig. 11c, d), width of the reaction zone clearly increases and quantity of α_1 phase, containing mostly Fe and Si, also increases. Width of the reaction zone is similar on both sides of the joint, slightly above 20 μm just at the lower edge of the steel sheet.

Figure 12 shows microstructure of the braze weld reaction zone and its surroundings from Fig. 11a, with use of the contrast material from the place where the reaction zone is ca. 18- μm thick. Table 4 shows results of quantitative microanalysis for the reaction zone and its surroundings in the braze weld from Fig. 12. It can be concluded on the grounds of these and the previous (Table 3) results that steel undergoes slight partial melting and creates with the liquid filler metal a non-homogenous liquid solution with high Fe concentration in vicinity of steel, and Si from the filler metal diffuses to the zone rich in Fe. When the joint is cooled down, various mixtures of the solutions Fe(Si) and Cu(Fe,Si) are created in the braze weld.

It can be stated on the grounds of EDS microanalysis that the reaction zone is composed of the matrix rich in Fe and Si (dark phase) and particles of the phase rich in Cu (bright

Fig. 12 Microstructure of the braze weld reaction zone and its surroundings from Fig. 11a. SEM (material contrast). Points of EDS analysis are marked. Primary beam energy $E_0=20$ keV

phase); its small particles occur on the entire width of the reaction zone and big particles—on the weld side. Results of microanalysis of the phase rich in Fe and Si in the reaction zone are disturbed by tiny particles of the phase rich in Cu. Concentration of copper is overstated in relation to the other elements. It can be supposed that, in the zone, the

Table 4 Results of quantitative microanalysis for reaction zone and braze weld from Fig. 12

Point No	Location	%	Element			
			Cu	Fe	Si	Mn
1	Reaction zone, dark phase	wt%	15.1	79.1	5.8	–
		at.%	12.7	76.1	11.1	–
2	Reaction zone, dark phase	wt%	7.0	87.1	5.9	–
		at.%	5.9	82.9	11.2	–
3	Reaction zone, bright phase	wt%	89.9	8.8	0.7	0.7
		at.%	88.0	9.8	1.5	0.7
4	Braze weld, bright phase	wt%	93.3	5.0	1.0	–
		at.%	91.4	5.6	2.2	–
5	Braze weld, bright phase	wt%	94.5	3.6	1.1	0.8
		at.%	92.6	4.0	2.5	0.9
6	Braze weld, dark phase	wt%	7.3	84.3	8.3	–
		at.%	6.0	78.6	15.4	–

Fig. 13 Microstructure of the braze weld reaction zone and its surroundings from Fig. 11c. SEM (material contrast). Linear distribution of elements along the red line: Fe—white line, Cu—yellow line, Si—blue line

intermetallic phase α_1 (or α_2 —concentration of Si is close to the limit between both phases) is present, containing precipitates of the solution of Fe, Si and Mn in Cu (ca. 90 wt% Cu). Manganese occurs mainly in the precipitates; its concentration in the phase α_1 is below the spectrometer detectability. The braze weld is composed of the matrix being a solution of Fe, Si and Mn in Cu and particles rich in Fe and Si; some of them contain tiny precipitates of the phase rich in Cu. Concentration of copper was found higher in the braze weld than in the reaction zone. The precipitates in the braze weld contain over 15 at.% Si, which indicates the phase α_1 .

Figure 13 shows linear distribution of elements in the reaction zone and its surroundings from Fig. 11c, in the place where the reaction zone is 16–24- μ m thick. Like in the previous location, the zone contains Cu-rich particles—tiny on the entire surface and large on the braze weld side. Quantitative analysis for the specimen from Fig. 13 indicates that concentration of silicon in the reaction zone increases towards the braze weld, reaching over 17 at.% near the zone, i.e. 2 times more than near the steel. Compositions of the Cu-rich precipitate and of the braze weld are similar like in the previous location. Concentration of Si in the reaction zone reaches ca. 25 at.%, i.e. the upper limit for the phase α_1 . An increase of Si concentration in the reaction zone towards the braze weld is also visible in Fig. 14 showing distribution maps of the elements.

7 Mechanical testing of braze welded joints

Specimens for the static tensile test were prepared according to EN ISO 4136:2013-05 and EN 12797:2002/A1:2005. From the braze welded joints, specimens 20-mm wide were cut out, and then milled to 12 mm in the gauge length. Tensile tests of the braze welded joints were carried out on a hydraulic universal testing machine. The measuring range was up to 40 kN and travelling speed of the traverse was 2 mm/min. Average tensile strength of the braze welded joints was 204.5 MPa (average from 2 tests, because of limited quantity of the tested material). The fracture mechanism was cohesive, since the specimens were failed within the braze weld or partially within the braze weld and the reaction zone (Fig. 15).

Fig. 14 SEM image from Fig. 13 (a) and distribution maps of Cu (b), Fe (c) and Si (d) on surface of the braze welded joint

The obtained results are much better than those obtained for the same type joints made by flame ($C_2H_2-O_2$) braze welding with the use of coated brass filler wire grade 18XFC [15], as shown in Fig. 16. Tensile strength of the flame brazed joints was in average 145 MPa (in the range of $139 \div 150$ MPa) and the fracture mechanism was adhesive—the braze weld was torn-off from the 10CrMo9-10 steel on side surface of the chamfered tube groove.

Results of the technological bending test are also significantly diversified and indicate that, in the considered application case, better results can be obtained by the arc-brazing CMT technology (Fig. 17). The bending tests were continued to the moment when the first cracks appeared in the joint or in its vicinity. In the case of the CMT brazed joints, the bending angle was over 4 times bigger, equal to 45° , which is a very good result, taking into account significant thickness of the joined elements and the applied CMT brazing technology.

Vickers hardness measurements in the braze welded joint of Cu-lined 10CrMo9-10 steel tube were made acc. to EN ISO 6507-1:2007, using a tester Zwick 321 under the indenter load of 10 kG (98.07 N) for 15 s. Hardness of the base material (steel 10CrMo9-10) ranged within 157–168

HV 10 and hardness in the heat-affected zone was 181 HV 10. The braze weld created of the filler metal CuSi3 is soft; its hardness is ranging within 86–92 HV 10. In addition, hardness at 50 G load was measured in the reaction zone due to its small width. The reaction zone is the hardest place in the brazed joint, with its hardness equal to 491.1 HV 0.05. The softest zone is matrix of the braze weld CuSi3, with its hardness equal to 134.6 HV 0.05, see Fig. 18.

8 Conclusions

Bonding copper with steel is not an easy process and is problematic in the case of welding, where, as a result of significant mixing of copper with steel, hot cracks occur. Application of the low-energy CMT method for braze welding of boiler 10CrMo9-10 steel lined with copper makes it possible to obtain joints with good quality, highly aesthetic and with no spattering. Even if the braze welding process was applied, intensive diffusion processes proceeded and slight partial melting of the base material (10CrMo9-10 steel) occurred, especially in the weld face and root areas. As a result, a ca. 24-µm-wide reaction zone at the boundary between steel

Fig. 15 Exemplary fractures of the joints after tensile test: cohesive fracture in the braze weld (a) and cohesive fracture in the braze weld and in the reaction zone (b)

Fig. 16 Tensile strength of braze welded joints of copper-lined 10CrMo9-10 tube

and the braze weld was created. The reaction zone consists mostly of phase α_1 being the ordered solution $\text{Fe}_\alpha(\text{Si})$. This is a hard and brittle phase showing the highest hardness in the braze weld (491.1 HV 0.05); however, no cracks were observed in this phase. Tiny precipitates of α_1 phase occur in the braze weld, but they can not significantly affect properties of the joint. Mechanical properties of CMT arc braze welded joints are good and much higher than those of the same type flame braze welded joints. Tensile strength of the joints is 204.5 MPa and the cohesive fracture is located within the soft braze weld. Application of the CMT arc braze

Fig. 17 Joints of Cu-lined 10CrMo9-10 tube after technological bending test: flame braze welded (a) and CMT arc-braze welded (b)

welding technology eliminates or highly restricts the risk of hot cracking, both in steel as a result of copper diffusion from the filler metal along grain boundaries, and in the braze weld itself.

Fig. 18 Hardness HV 0.05 in braze weld of the 10CrMo9-10 steel tube, made using bronze filler metal CuSi3

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