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THE EFFECT OF RESIDUALS ON THE SUSCEPTIBILITY TO SURFACE CRACKING AND HOT DEFORMABILITY OF PLAIN CARBON STEELS

VPLIV OLIGOELEMENTOV NA POJAV POVRŠINSKIH RAZPOK IN DEFORMABILNOST V TOPLEM STANJU NIZKOOGLJIČNIH JEKEL

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The susceptibility to surface cracking, deformability and enrichment of the austenite grain boundaries with residuals during heating carbon steel with an increased content of Cu (0.24-1.10%), Sn (0.020-0.105%) and Sb (0.003-0.042%) were investigated using a bending test, a hot-torsion test and electron-probe microanalysis. Before testing, the steels were heated for three hours in a furnace with an air atmosphere at 900-1250 °C.

The differences in the enrichment of Cu and Sn at the metal/oxide interface and the surface hot shortness of steels with different residuals' contents within the temperature range 900-1250 °C were investigated. We found that the surface-layer enrichment with residuals and the propensity to form surface cracks during hot deformation are dependent on the temperature. A residuals-enriched subscale layer cannot affect the workability measured with a hot-torsion test although the number of revolutions to fracture at high temperatures depends strongly on the average content of residuals in the steel, especially the content of tin.

Key words: carbon steel, surface hot shortness, deformability, surface cracks, surface-layer, residuals, bending test, hot-torsion test

Ugotovljali smo nagnjenost k nastanku površinskih razpok, deformabilnosti pri toplem valjanju in obogatitvi meja avstenitnih zrn oligoelementov pri segrevanju nizkoogljičnih jekel s povišano vsebnostjo bakra (0,24 - 1,1 %), kositra (0,021 - 0,105 %) in antimona (0,003 - 0,042 %). Jekla smo segrevali pri temperaturi 900 - 1200 °C 3 ure v elektrouporovni peči v zračni atmosferi.

Razprava se nanaša na rezultate raziskav kopičenja oligoelementov v površinskih plasteh pod oksidno skorjo, površinske razpoke in deformabilnost v toplem stanju pri segrevanju pri različnih temperaturah. Obravnavali smo razliko med intenziteto kopičenja Cu in Sn na mejah kovina/oksid in površinske razpoke med jeklom z različnimi vsebnostmi oligoelementov v intervalu 900 - 1200 °C. Ugotovili smo, da je obogatitev površinskih slojev z oligoelementi zelo odvisna od temperature segrevanja.

Analogno se spreminja nagnjenost k nastajanju površinskih razpok pri topli deformaciji, vendar pa kopičenje oligoelementov pod oksidno skorjo bistveno ne vpliva na deformabilnost pri toplem valjanju, število upogibov do zloma pri visokih temperaturah pa je odvisno od njihove povprečne vsebnosti v jeklu - predvsem kositra.

Ključne besede: ogljična jekla, površinska lomljivost, deformabilnost, razpokanost površine, oligoelementi

1 INTRODUCTION

The production and processing of electrical steels can be problematic as a result of the presence of residuals such as copper and tin. Antimony and arsenic can cause similar problems but these elements are less likely to be present in steel. All of these elements, which are referred to as residuals, originate from steel scrap. They have less affinity for oxygen than iron and cannot be removed by conventional refining techniques. Residuals can cause surface cracks during hot deformation [1,2] or continuous casting [3,4], and can have an even greater effect on other features of steels [1,5].

When steel is heated the copper, which is a more noble metal than iron, becomes enriched in the vicinity of the metal/oxide interface [6] due to selective surface oxidation. If the copper concentration in a thin subscale layer becomes higher than the solubility limit in γ-iron (~8 % Cu) [7], a new copper-enriched zone with a low melting point will be formed. According to the FeCu [8] phase diagram, this phase becomes liquid at 1094 °C (the peritectic point). The liquid phase that is enriched in copper penetrates along the austenite grain boundaries, weakens the bonds and causes inter-crystalline surface cracks as a result of tensile strain. The copper-enriched phase usually contains other elements in solution such as tin, antimony and arsenic [9], which are also more noble than the iron. These elements have the same negative effect as copper in terms of the formation of surface cracks during high-temperature deformation. The content of these elements in the copper-enriched liquid phase depends primarily on their average concentration in the steel and the heating temperature.

Table 1: Chemical composition (wt%) of the steel used for remelting

C	Si	Mn	P	S	Cu	Cr	Ni	Mo	Al	As	Sn	Sb	Pb
0.14	0.16	0.35	0.013	0.027	0.18	0.09	0.09	0.03	0.004	0.010	0.020	0.003	0.004

Table 2: Content of copper, tin and antimony (wt.%) in experimental steels

Ch. element	Designation of experimental steels										
	A	B	C	D	E	F	G	H	I	J	K
Cu	0.24	0.45	0.51	0.81	1.11	0.18	0.44	0.61	0.56	0.56	0.62
Sn	0.021	0.020	0.021	0.020	0.022	0.105	0.098	0.082	0.042	0.072	0.042
Sb	0.003	0.002	0.004	0.002	0.002	0.004	0.003	0.004	0.002	0.042	0.027

The levels of residuals in electrical steels are restricted by the quality criteria, which are tending to become more demanding and strict, however, levels in continuous cast steel are increasing due to the purchase of more scrap with higher levels of residuals. The quality of steel scrap is declining owing to a significant share of it containing copper and tin from baled car bodies. In circumstances when a low-quality scrap has to be used, a detailed study of the technological factors connected to the negative effects of residuals during high-temperature steel processing is necessary. In this paper the result of an investigation of a series of laboratory-made plain carbon steels with various contents of copper; copper and tin; and copper, tin and antimony, is discussed. Along with a study of surface shortness at different temperatures, the enrichment of the surface layer and austenite boundaries was also investigated.

2 EXPERIMENTAL

A series of experimental steels for testing the effect of residuals on hot ductility was prepared in a laboratory induction furnace by remelting continuously-cast plain carbon steel billets with the chemical compositions given in **Table 1**.

By melting and after-alloying, eleven steel grades with different contents of copper; copper and tin; and copper, tin and antimony; were prepared (**Table 2**). Two 45-kg ingots of each steel were produced with square cross-sections. One ingot was hot forged into 80×80-mm bars and then hot-rolled into Φ12-mm round bars.

To estimate a steel's susceptibility to surface cracks we used a hot bending test. Rolled bar samples, 200-mm long, were annealed in an electrical resistance furnace for 3 hours at 900-1250 °C in air. After annealing the samples were removed from the furnace and immediately bent by 90° in a unit designed for this purpose.

The number of cracks on the bent part of the test bar, which was determined by simple inspection, was used as an indication of the steel's susceptibility to surface cracking, and each steel was graded in the range 0 to 6: 0 - free of cracks, 1 - "orange skin" type rough surface with a small number of light cracks, 2 - many light cracks, 3 - many light cracks with a small number of long cracks, 4 - many long cracks, 5 - clearly marked long and wide cracks, 6 - severely torn surface with deep uneven cracks.

The workability of the steel was tested with the hot-torsion method using a SETERAM torsion machine. All test pieces were cut from the rolled bars and machined to a diameter of 6 mm and a gauge length of 50 mm. After machining, some of the test bars were pre-annealed in an electric furnace in the temperature range 900-1200 °C in air for 3 hours and then cooled in air, while another series was tested in the as-hot-rolled condition, without any pre-annealing before the hot-torsion test. As a part of the hot-torsion test the test bars were reheated in air according to the following regime: 1 °C/s heating rate to the testing temperature (900-1200 °C) and then held at temperature for 15 minutes. The specimens were then twisted continuously at a strain rate of 1.0 s^{-1} until they fractured. The number of revolutions to fracture was used as a quantitative measure of the samples' hot workability.

The enrichment of copper, tin and antimony in the subscale layer and at the austenite grain boundaries as well as their dendritic segregation were investigated using a JEOL JCXA-733 electron-probe X-ray microanalyzer.

3 RESULTS AND DISCUSSION

3.1 Surface cracking of the steel

In **Figure 1** the level of surface cracking versus the heating temperature of the A, B, D, and E steels containing different amounts of copper but the same amounts of tin and antimony is shown. The increased copper content resulted in severe surface cracking in the range 1000-1200 °C, with the maximum shortness occurring at higher temperatures. For the A and B steels the most severe cracks occurred with the specimens annealed at 1100 °C, while for the D and E specimens the maximum cracking was found for the samples annealed at 1150 °C. All five steel specimens annealed at 1250 °C withstood bending with no cracking and only for the D and E steels was the "orange skin"-type rough surface observed.

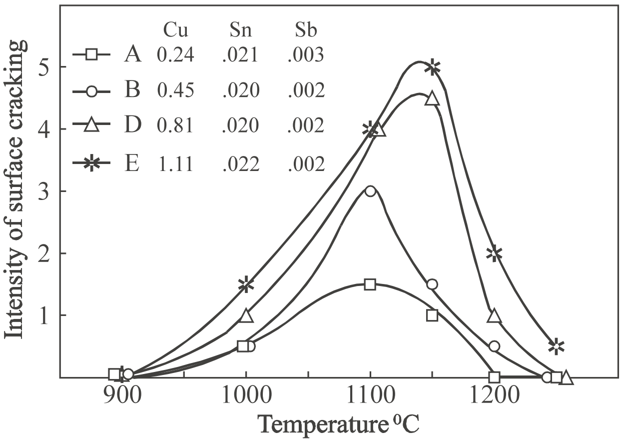

Figure 1: Intensity of surface cracking in dependance on heating temperature for the A, B,D, and E steels withg different content of Cu and the constant content of Sn and Sb

Slika 1: Odvisnost površinskih razpok od temperature segrevanja za jekla A, B, D, i E, ki imajo različno vsebnost Cu in enako vsebnost Sn in Sb

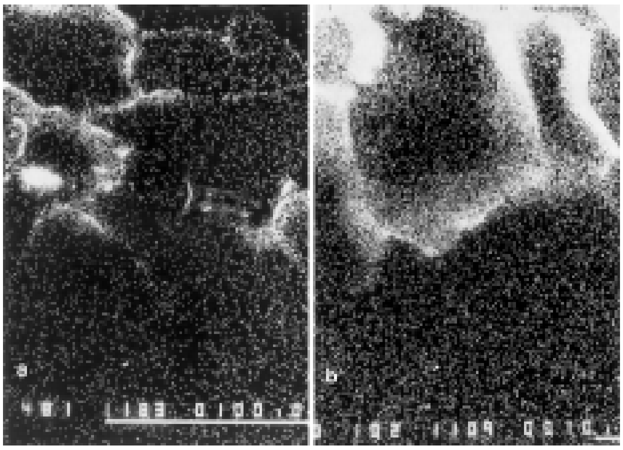

Figure 3: Cu enrichment in surface layer and penetration of liquid phase along austenite grain boundaries: a) B steel at 1100 °C; b) D steel at 1150 °C

Slika 3: Obogatitev Cu v površinski plasti in prodor tekoče faze po kristalnih mejah avstenita za jeklo: a)1100 °C; b) 1150 °C

In the critical temperature range 1100-1150 °C the surface shortness increased rapidly with a corresponding increase in the copper content up to 0.60%. Any further increase in the copper content had only a negligible effect on the shortness (**Figure 2**).

The analyses in the electron-probe microanalyser showed that the surface shortness is directly connected to the Cu enrichment in the subscale layer. Both the annealing temperature and the copper content of the steel had a crucial influence on the Cu enrichment at the metal/oxide interface, the formation of the liquid phase and its penetration along the austenite grain boundaries. With an increase of the annealing temperature the surface Cu enrichment increased too and reached its maximum value at a critical temperature. After this it decreased and gradually disappeared at a still higher temperature. During annealing at 900 °C only the D and E steels exhibited a measurable content of Cu on the surface, however, after heating at 1000 °C the Cu enrichment at the metal/oxide interface was evident on all five steel specimens. The greatest Cu enrichment in the surface layer and the deepest penetration of the liquid phase along the austenite grain boundaries of the A, B and C steels occurred at 1100 °C (**Figure 3a, 3b**). The surface Cu enrichment decreased and then gradually disappeared at higher annealing temperatures: at 1200 °C for steel A and at 1250 °C for steel E (**Figure 4**).

The Cu enrichment in the surface layer is a result of the selective iron oxidation, and the changes in the Cu content are due to the competition between surface oxidation and the rate of diffusion of copper into the metal and along the grain boundaries (**Figure 5**) as well as the occlusion of metal particles within the scale layer [10,11] (**Figure 4**). Thus, the copper content in the enriched layer at a particular temperature depends on its content in the steel, on the surface oxidation rate and on the

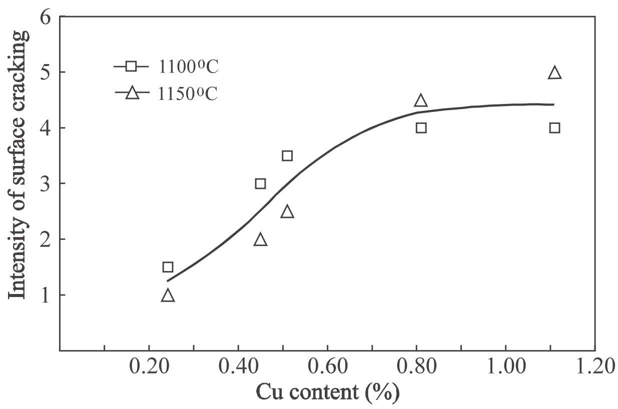

Figure 2: Effect of Cu content on surface shortness intensity at critical temperatures 1100-1150 °C

Slika 2: Vpliv vsebnosti Cu na pokljivost površine pri kritični temperaturi 1100-1150 °C

Figure 4: Absence of Cu enrichment in surface layer at 1250 °C for steel E

Slika 4: Odsotnost obogatitve Cu na površinski plasti pri temperaturi 1250 °C za jeklo E

Figure 5: Cu distribution along boundaries and into austenite grains in surface layer at 1200 °C for steel D

Slika 5: Porazdelitev Cu na mejah in na avstenitnih zrnih pri temperaturi 1200 °C za jeklo D

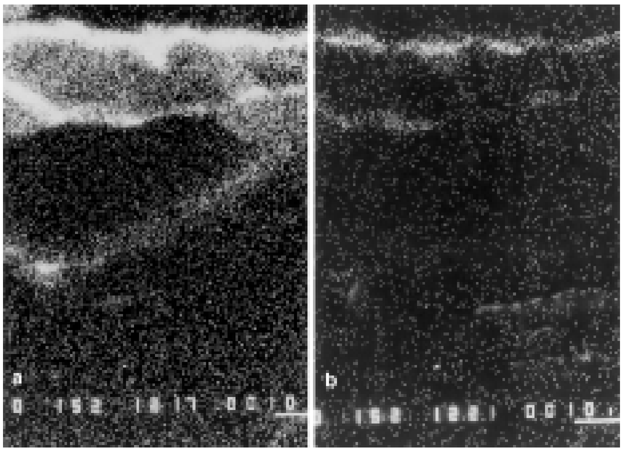

Figure 7: Distribution of Cu and Sn in subscale layer in G steel at 1150 °C

Slika 7: Porazdelitev Cu in Sn pod škajo za jeklo G pri 1150 °C

copper diffusion rate in the austenite, as well as its occlusion within the oxide layer. All these processes are faster at higher temperatures, while the copper leaving the enriched layer prevails at moderately high temperatures.

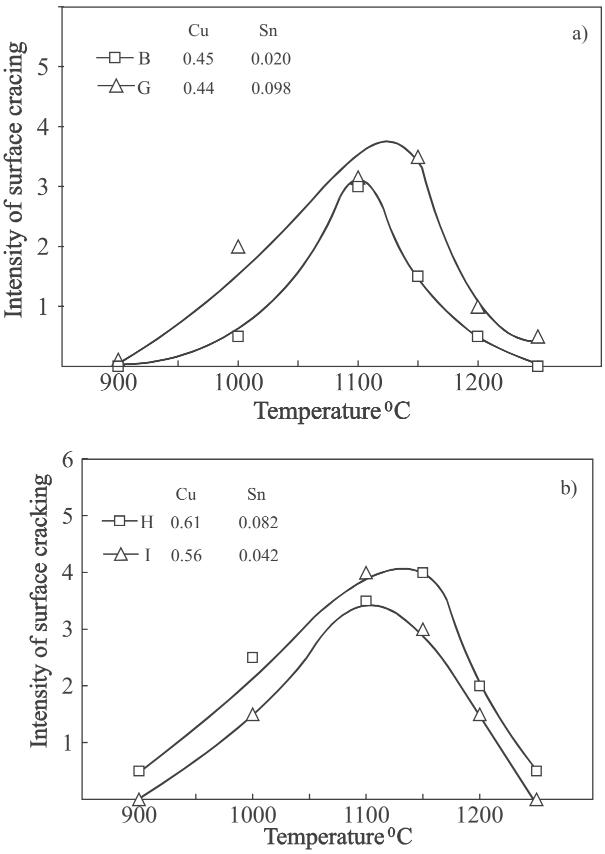

Figure 6: Intensity of surface cracking as function of temperature in: (a) B, G and (b) H, I steels

Slika 6: Intenziteti razpokanja v odvisnosti od temperature: a) jekla B,G in b) H, I

The increased tin content in the steel intensifies the effect of copper on the surface shortness. **Figures 6a and 6b** illustrate the levels of surface cracking as a function of temperature in the B, G, H and I steels. The high tin content in the G steel (**Figure 6a**) produced a larger temperature shortness area at both higher and lower temperatures, compared to the low-tin B steel. Similar test results were obtained for the H and I steels (**Figure 6b**) with almost the same copper but different tin content.

For the G, H and I steels, with the high copper and tin contents, annealing at 1000-1200 °C gave rise to enrichment of both elements in the subscale layer with a maximum at 1150 °C (**Figure 7**). Tin reduces the solubility of Cu in γ-iron, it is dissolved in the Cu-enriched phase and decreases its melting point [6]. The enlargement of the surface shortness area at higher temperatures (**Figure 6**), due to the content of tin, probably resulted in the lower copper diffusion rate, also

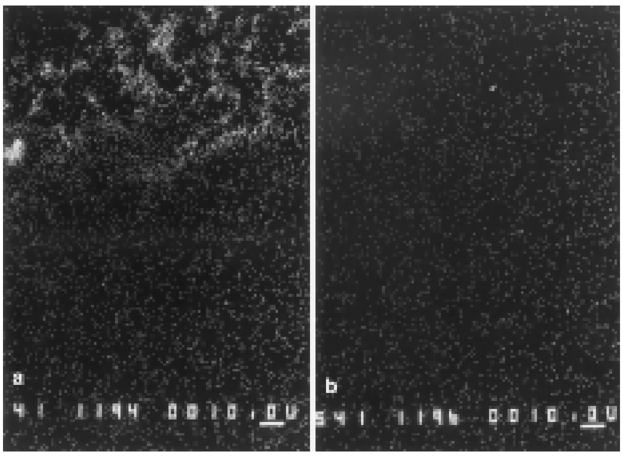

Figure 8: Distribution of Cu and Sn in subscale layer for F steel at 1100 °C

Slika 8: Porazdelitev Cu in Sn pod škajo za jeklo F pri 1100 °C

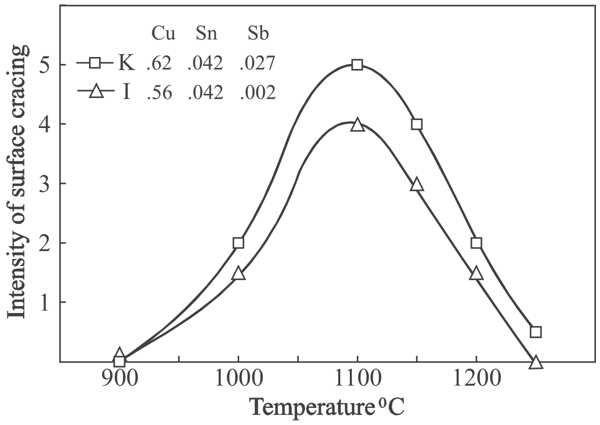

Figure 9: Surface crack intensity changes in function of temperature for I and K steels
Slika 9: Sprememba intenzitete površinskih razpok v funkciji temperature za jekli I in K

due to the presence of tin [12]. The enlargement of the shortness area at lower temperatures under the influence of the tin can result from the lower melting temperature of the Cu-enriched liquid phase [6] and the lower Cu solubility in iron, which leads to a liquid phase with a lower Cu content at the surface.

Unlike the G, H and I steels, which have high Cu and Sn contents, the F steel with its low copper (0.18%) and high tin (0.105%) contents did not exhibit surface shortness during hot deformation. All the test bars withstood bending to 90° without any cracking. Only the test bar annealed at 1100 °C showed a rough, "orange skin"-type surface. On this bar we observed an enrichment of the copper, but no enrichment of the tin (**Figure 8**). It is likely that the tin in the

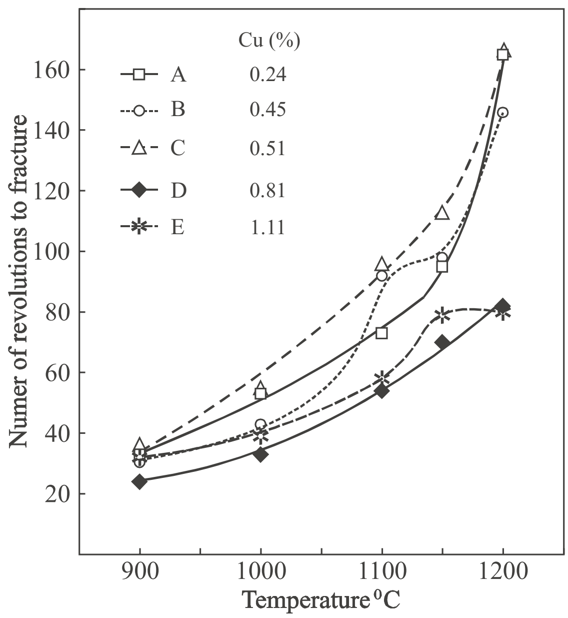

Figure 10: Number of revolutions to fracture in dependence test temperature for the A - E steels with different copper contents
Slika 10: Število vzvojev do loma za jekla od A do E z različno vsebnostjo Cu v odvisnosti od temperature preskusa

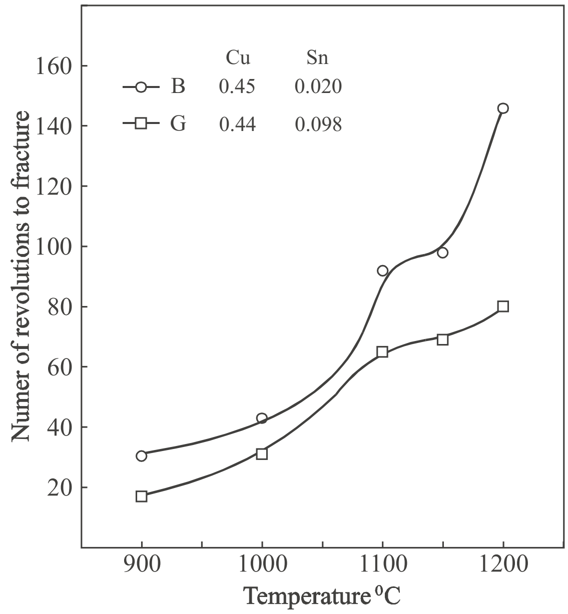

Figure 11: Number of revolutions to fracture in depedence on the temperature in steels B and G with identical content of copper and different content of tin
Slika 11: Število vzvojev do loma za jekla B in G z identično vsebnostjo Cu in različno vsebnostjo Sn

low-Cu-containing steel moves relatively fast by diffusion from a metal/oxide interface to the austenite so that during prolonged annealing its negative effect on the appearance of the surface defects disappears.

In **Figure 9** the surface-crack intensity change as a function of the temperature is shown for I and K steels, which had the same Cu and Sn but different Sb contents. The high Sb content produced an increased surface shortness in the range 1000-1200 °C. Like Sn, Sb is enriched in the surface layer that is already enriched in Cu.

3.2 Workability during hot torsion

In **Figure 10** the number of revolutions to fracture, and its dependence on the temperature for the A-E steels with different copper contents (**Table 2**) is shown. At 900 °C we found no effect of the copper content increase on the workability of the steel. However, at the higher test temperatures, low-copper (<0.60%) steels had a much higher ductility than the high-copper (0.81 and 1.11%) steels.

Tin affected the steel's hot-torsion workability very significantly, as shown in **Figure 11**, where the number of revolutions to fracture as a function of temperature in steels with identical amounts of copper but different amounts of tin is given. The negative effect of tin increases with higher temperatures.

Figure 12 illustrates that the number of revolutions to fracture depends on the copper and tin contents and the test temperature. The upper curve relates to the low-tin A-E steels, while, the lower curve shows the

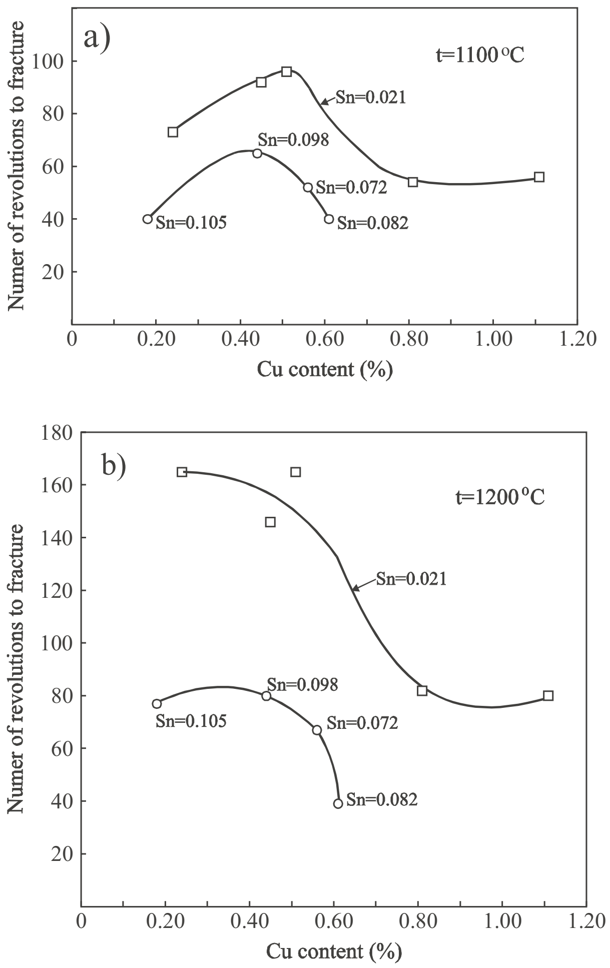

Figure 12: Number of revolutions to fracture by different copper and tin contents at different test temperature: a) 1100 °C; b) 1200 °C

Slika 12: Število vzvojev do loma pri različni vsebnosti Cu in različni temperaturi: a) 1100 °C; b) 1200 °C

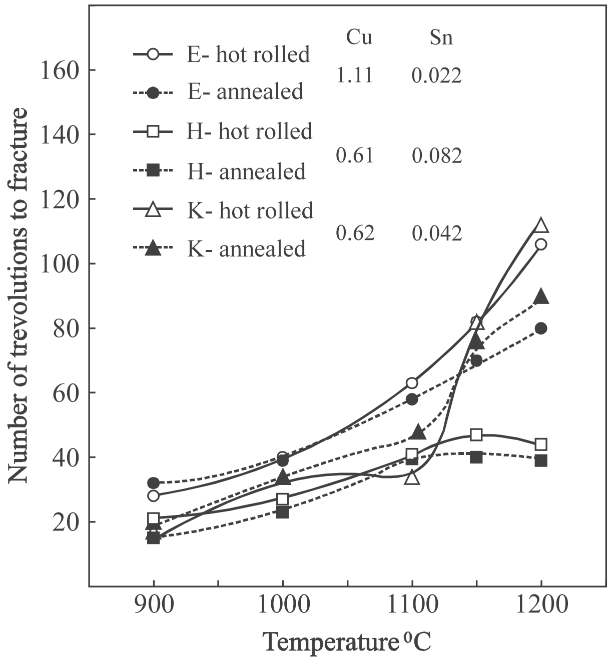

Figure 13: Number of revolutions to fracture in dependence on temperatures for the E, H and K steels in annealed and as hot rolled conditions

Slika 13: Število vzvojev do loma v odvisnosti od temperature za jekla E, H in K v analognih razmerah toplega valjanja

influence of tin. High-tin (0.042-0.105%) specimens withstood fewer revolutions than the low-tin steels. The negative influence of copper on the workability, in presence of tin, becomes apparent at a copper content below 0.60%. It is interesting that the low-copper and high-tin F steel, which does not show any surface shortness during hot bending, does have a low workability during hot torsion.

Figure 13 shows the number of revolution to fracture for different temperatures for the E, H and K steels. Pre-annealing before the hot-torsion test had almost no effect on the workability, except for steel E at the higher temperatures: 1150 °C and 1200 °C. The test results indicate that the very negative influence of tin on the steel's hot workability is not the result of tin enrichment in a subscale layer. Tin has a tendency, like sulphur [13], to macro-segregate during steel solidification; SEM linear-profile test results also showed a strong dendritic segregation, which could decrease the steel's hot workability.

4 CONCLUSIONS

The copper enrichment at a metal/oxide interface increases with temperature to a maximum at 1100-1150 °C. Above this limit it drops gradually and disappears at 1200-1250 °C. The increased Cu content of the steel increases the temperature of the maximum Cu enrichment and the temperature when the enriched layer disappears. The shortness of steel surface changes analogously to the level of Cu enrichment at the metal/oxide interface.

In high-Cu-content steel, tin is enriched in the subscale layer and extends the range of the surface shortness towards higher and lower temperatures. In low-Cu-content steel, tin is not enriched at the metal/oxide interface, and therefore the surface cracks do not occur during hot bending.

Similarly to tin, antimony is enriched in the Cu-enriched surface layer. It intensifies the susceptibility to surface cracking and enlarges the temperature range of the surface shortness.

An increased Sn content decreases the workability during hot torsion, especially at high temperatures, while copper's negative effect only shows up at high concentrations (>0.6-0.7%). Unlike surface shortness, surface-layer enrichment with residuals does not affect the hot-torsion workability.

5 LITERATURE

[1] D. A. Melford: Residuals Additives and Materials Properties, The Royal Society, London, 1980, 90-103

[2] B. Mintz, ISIJ International, 39 (**1999**) 9, 833-855

[3] M. H. Barden, G. D. Funnel, A. G. Whitoker and J. M. Young: Solidification Casting of Metals. The Metals Society, London, 1979, 279-286

[4] M. M. Wolf, ISM, april 2000, 58-60

[5] W. Leslie: The Physical Metallurgy of steels, McGraw-Hill International Book Company-Casaido Printing Co. LTD Tokyo, 1982, 236-242

[6] F. Vodopivec, O. Kürner, J. Rodič, M. Vuk, Metalurško posvetovanje Portorož (1975)

[7] A. Kveder, Poročilo Metalurškega inštituta, Ljubljana, MI 381-405, november 1986

[8] E. Hudremont: Special'nye stali, Izdateljstvo Metallurgija, Moskva, 1966

[9] E. Anelli et al., Proc. of 34th Mech. Working and Steel Processing Conf., ISS-AIME, Warendale, PA, 30 (**1993**), 399-407

[10] Y. Pohsafe, C. Cuche: Proc. Copper in Steel, INCRA, New York, 1983, 9.1-9.24

[11] M. Djurović, B. Perović, M. Andjelić, K. Kovačević, Metal 2000, 9th International Metallurgical Conference, Ostrava, 2000

[12] D. A. Porter, K. E. Eassterling: Phase Transformations in Metals and Alloys, VNR 1981, 102

[13] B. Perović et al., Zbornik radova Kolokvijuma o livarstvu, Budva, 1998