



## Effect of segregations on mechanical properties and crack propagation in spring steel

B. Žužek, M. Sedlaček, B. Podgornik

*Institute of Metals and Technology, Lepi pot 11, SI-1000 Ljubljana, Slovenia*  
*borut.zuzek@imt.si*

**ABSTRACT.** Considerable efforts have been made over the last decades to improve performance of spring steels, which would increase the service time of springs and also allow vehicles weight reduction. There are different possibilities of improving properties of spring steels, from modifying the chemical composition of steels to optimizing the deformation process and changing the heat treatment parameters. Another way of improving steel properties is through refining the microstructure and reducing amount of inclusions. Therefore, the focus of the current investigation was to determine the effect of more uniform and cleaner microstructure obtained through electro-slag remelting (ESR) of steel on the mechanical and dynamic properties of spring steel, with special focus on the resistance to fatigue crack propagation. Effect of the microstructure refinement was evaluated in terms of tensile strength, elongation, fracture and impact toughness, and fatigue resistance under bending and tensile loading. After the mechanical tests the fracture surfaces of samples were analyzed using scanning electron microscope (SEM) and the influence of microstructure properties on the crack propagation and crack propagation resistance was studied. Investigation was performed on hot rolled, soft annealed and vacuum heat treated 51CrV4 spring steel produced by conventional continuous casting and compared with steel additional refined through ESR. Results shows that elimination of segregations and microstructure refinement using additional ESR process gives some improvement in terms of better repeatability and reduced scattering, but on the other hand it has negative effect on crack propagation resistance and fatigue properties of the spring steel.

**KEYWORDS.** Spring steel; Crack propagation; Electro-slag remelting; Segregations; Fracture toughness.

### INTRODUCTION

Increasing demands of industry, especial automotive industry on performance improvement of steels put a lot of pressure on the steel and steel parts producers. The pressure is in weight reduction and cost savings what governs the producers in new design concepts and further material development. Weight reduction is very important because it reduces costs and even more importantly it reduces fuel consumption and CO<sub>2</sub> pollution in automotive industry. One of the biggest fuel consumers and polluters are trucks, where redesign and use of lighter high strength leaf springs can bring considerable benefits. Beside lower weight of springs, reduced dimensions also allow increased flexibility in design, enabling more room for additional safety components. Traditional parabolic leaf springs used in suspension systems of truck front axles are usually made of two leafs, and serve two main purposes: support the weight of the trailer and also



provide the spring function in the suspension system. With the new spring designs, eventually aiming at a single leaf solution, savings of over 20% of the total weight of the spring can be expected. However, this would also lead to about 10-15% increase in spring maximum stress, which requires better spring steel with ultimate tensile strength of over 2.000 MPa [1,2].

Considerable efforts have been made over the last decades to develop high strength spring steels to meet the needs for weight and cost savings in the automotive industry [3]. Improved properties of spring steel can be achieved through precise control of chemical composition of steel, optimum heat treatment, micro-alloying, thermomechanical treatment and shot-peening [3-12]. For the martensitic steels the best possibility for strength improvement are achievable through lowering the austenizing and tempering temperature, which leads to the increase of ultimate tensile strength but on the other hand it reduces the steel ductility and toughness [8,13]. However, in the case of springs improvement in strength should not degrade other spring steel properties such as formability and fatigue resistance [6]. Another way of improving spring steel strength is through grain refinement [5,6,14], mainly based on micro-alloying and thermomechanical treatment. The addition of certain alloying elements has been reported to effectively improve spring steel strength as well as sag resistance [3]. Si of up to 2% was found to improve properties through the refinement of tempered carbides obtained by retardation of  $\epsilon$ -carbide conversion to cementite during tempering [3,15]. The addition of Nb and V, has also been reported beneficial due to the precipitation and dispersion of fine micro-alloyed carbonitrides [9]. Further improvement in tensile properties can be obtained through deformation of austenite prior to quenching [8]. Improvement is related to refined austenite grains and to the austenite grain substructure. Those conditions reflect into finer structured martensite, i.e. refined block size, with no or refined carbides at the prior austenite grain boundaries.

Most commercial steels, including spring steels contain impurities that significantly influence the ductility and toughness, with the loss in properties being dependent on the impurity element concentration [16]. The well-known embrittlement phenomena observed around 350°C is such an example where grain boundary segregation of impurity element together with carbide films at grain boundaries deteriorate the mechanical properties of the steel [17]. However, there are also non-metallic inclusions, which depending on the type and size, may further degrade spring steel properties [18-20].

In the case of tool steels microstructure refinement and properties improvement can be achieved through utilization of electro-slag remelting (ESR) [21]. Uniform, relatively rapid solidification in ESR with highly reactive slag leads to an improvement in the chemical uniformity and uniformity of macrostructure, greatly improved cleanliness and reduced segregation tendency, removal of exogenous oxide inclusions and substantial sulphur reduction [21]. Microstructural features such as the eutectic cell size and the eutectic carbide particle size are also reduced. These features, beside other, result in improved hot workability and better ductility and fatigue properties of tool steels [22]. It has been shown, that the surface defects and inclusions have a huge effect on the fatigue life and on the fracture surface appearance, and it can also change the S-N property from duplex to single one or vice versa [23]. The condition of the surface of spring steel specimens, for instance just the grinding direction can change the fracture of the specimen from the surface-induced failure to the interior-induced failure [24].

The aim of the presented research work was to investigate the possibility of refining spring steel microstructure using electro-slag remelting and to determine its effect on the mechanical and dynamic properties of commercial 51CrV4 spring steel, with the focus on the resistance to fatigue crack propagation.

## EXPERIMENTAL

### *Material and heat treatment*

Material used in this investigation was commercial 51CrV4 spring steel produced by a conventional casting process used for the research in [25]. Two charges of steel (chemical composition of both charges is given in Tab. 1) were cast in a billets (180x180 mm). One charge, used as a reference and denoted CCC was directly hot rolled in strips (25x90 mm) and soft annealed, while the other, denoted ESR was first electro-slag remelted and then hot rolled and soft annealed under the same conditions as the first charge (CCC).

For each charge adequate specimens were taken from hot rolled and soft annealed stripes in the rolling direction and vacuum heat treated in a vacuum furnace with uniform high-pressure gas-quenching using nitrogen gas at a pressure of 5 bar. After heating (10°C/min) to the austenitizing temperature of 870°C, specimens were soaked for 10 min, gas quenched to a temperature of 60°C, and then single tempered for 1h at 300, 375 and 475°C, respectively.



Charge	C	Si	Mn	P	S	Cr	Al	Cu	V
ESR	0.53	0.29	0.94	0.010	0.003	1.07	0.025	0.18	0.16
CCC	0.54	0.33	0.97	0.012	0.007	1.10	0.006	0.17	0.17

Table 1: Chemical composition of CCC and ESR spring steel charges (mas. %).

### Mechanical testing

Mechanical properties involved tensile test, hardness measurement, as well as fracture and impact toughness analysis. Tensile test according to EN ISO 6892-1 A224 was performed on standard B10x50 mm specimens using universal testing machine Instron 8802, and all standard tensile test parameters were determined.

Fracture toughness measurement were performed on non-standard circumferentially notched and fatigue-precracked tensile-bar specimens (Fig. 1), designated CNPTB specimens [26]. Fatigue pre-crack of about 0.5 mm was created under rotating-bending loading before the final heat treatment [27]. Using Instron 8802 tensile-test machine and cross-head speed of 1.0 mm/min load at fracture was measured and fracture toughness  $K_{Ic}$  calculated using Eq. 1 [28]:

$$K_{Ic} = \frac{P}{D^{3/2}} \times \left( -1.27 + 1.72 \times \frac{D}{d} \right) \quad (1)$$

where  $P$  is the load at failure,  $D$  the outside non-notched diameter (10 mm), and  $d$  the diameter of the instantly fractured area. Eqn. (1) is valid for  $0.5 < d/D < 0.8$  and linearly elastic behavior, as displayed by all investigated specimens.

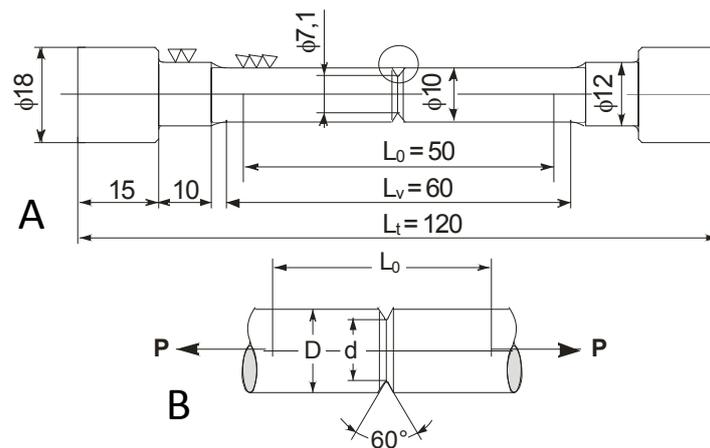


Figure 1: A) CNPTB specimen, circumferentially notched and fatigue-precracked (units in mm), B) detail on notch marked on Fig.1A.

Impact toughness was determined on Charpy V-notch specimens ( $KV_2$ ) according to standard EN ISO 148-1 using 300 J pendulum. Tests were performed at room and sub-zero temperatures of  $0^\circ\text{C}$ ,  $-20^\circ\text{C}$  and  $-40^\circ\text{C}$  in order to reveal ductile to brittle transition temperature (DBTT), carried out for specimens tempered at  $475^\circ\text{C}$ . Finally, the Rockwell hardness measurements (HRC) were performed on each cylindrical specimen using an Instron B2000 hardness testing machine.

### Dynamic testing

Fatigue properties were determined under bending as well as tensile-compression loading. Bending fatigue testing was performed on a Rumul Cracktronic resonant machine using standard  $10 \times 10 \times 55$  mm Charpy V-notch test specimens. The testing resonant stress frequency was 175 Hz using a sinusoidal waveform at a stress ratio  $R$  of 0.1. A constant amplitude bending stress, ranging between 275 MPa and 430 MPa was used. A criterion of specimen failure was a drop of inherent oscillation for more than 3%, where fatigue cracks occurred in a depth of up to 3 mm.

Fatigue testing under tension-compression loading were conducted in servo hydraulic Instron 8802 dynamic test machine under the conditions of  $R = -1$  and sine wave of 30 Hz. Tests were performed on polished hourglass-shaped fatigue specimens with a gauge length of 38 mm and neck diameter of 7.5 mm. Maximum tensile stresses were in the range between 500 and 780 MPa.

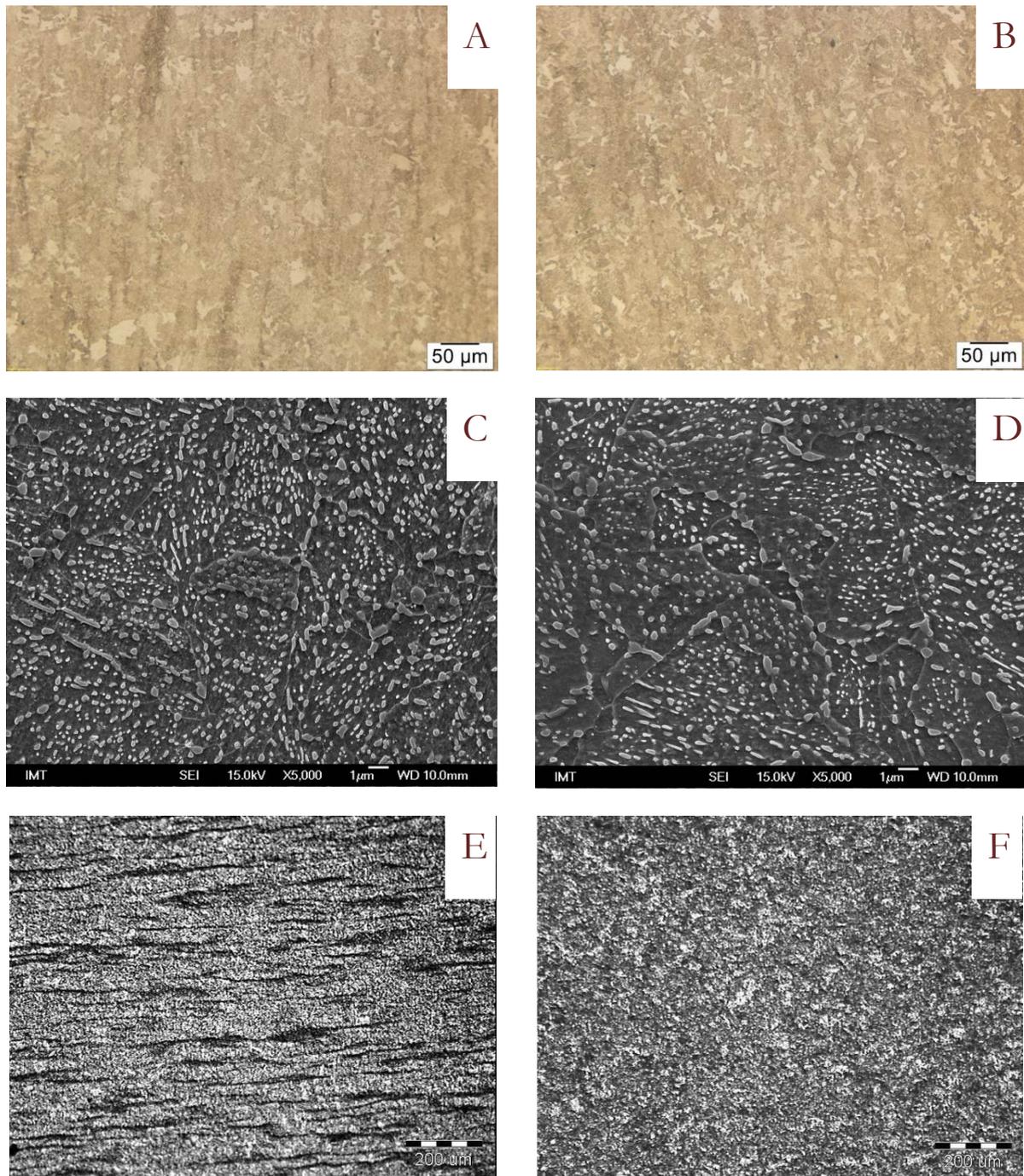


Figure 2: Microstructures of CCC specimens (A), (C) and (E) and ESR specimens (B), (D) and (F) of investigated spring steel in a longitudinal direction. All microstructures were revealed by Nital etching, with Figs. (E) and (F) over-etched to show the segregation phenomenon.

## RESULTS AND DISCUSSION

### *Microstructure*

**M**icrostructure of examined spring steel was investigated using light microscope (LM) as well as scanning electron microscope (SEM). Microstructure of conventionally cast (CCC) and electro-slag remelted (ESR) spring steel after hot rolling and soft annealing is shown in Fig. 2. Microstructure for both charges of steel consists of fine

carbides in ferrite phase (Fig. 2A and 2B). Carbides are mainly distributed along previous martensitic laths, with some of the carbides being slightly coarsened, mostly those along primary ferrite grain boundaries (Fig. 2C and 2D). No obvious difference in size and distribution of carbides between CCC and ESR specimens can be noticed. Nevertheless, in the case of ESR specimens more homogeneous microstructure was obtained without distinctive segregations and reduced number of non-metallic inclusions (Fig. 2E and 2F). Additional remelting of steel through ESR also slightly reduced the grain size, according to standard EN ISO 643 from G10 for CCC specimens to G11 for ESR specimens (Fig. 2C and 2D). Beside these, as indicated in Tab. 1, concentration of P and S, as well as other alloying elements has been slightly reduced by ESR. On one hand ESR resulted in refined microstructure of spring steel, but on the other hand ESR also caused the appearance of some  $\text{Al}_2\text{O}_3$  inclusions [27], otherwise absent in CCC specimens.

All three different tempering temperatures during heat treatment resulted in the martensitic microstructure, with very distinctive segregations being observed only in the case of CCC specimens.

### *Mechanical and toughness properties*

After vacuum heat treatment hardness of both CCC and ESR spring steel specimens was more or less the same. More uniform microstructure of ESR specimens with reduced number of inclusions increased yield and tensile strength. At the lowest tempering temperature (300°C) yield strength has been increased from 1735 MPa to 1755 MPa and tensile strength from 1960 MPa to 1990 MPa. For the highest tempering temperature (475°C) yield strength has been increased from 1410 MPa to 1420 MPa and tensile strength from 1480 MPa to 1485 MPa when using ESR refining method. However, the increase was less than 1.5%. On the other hand ESR greatly reduced scattering of results, and increased contraction for up to 40%, especially for the lowest tempering temperature.

More pronounced effect of ESR refinement was observed in the case of impact toughness. For CCC spring steel, tempered at 475°C impact toughness at 20°C was 20.6 J. By decreasing testing temperature to 0°C impact toughness of CCC spring steel dropped to 19.9 J and below -20°C even to 16.0 J. On the other hand, impact toughness for ESR specimens at 20°C increased to 22.0 J, which even at the lowest testing temperature of -40°C didn't drop below 21.5 J, thus showing much better resistance to low temperature brittleness.

Additional ESR refinement of investigated spring steel had practically no influence on the steel fracture toughness level. However, through more uniform microstructure and reduced number of inclusions scattering was reduced and fractured surfaces differ significantly (Fig. 3). In the case of CCC spring steel specimens fracture toughness after tempering at 300°C was  $25.8 \pm 1 \text{ MPa m}^{1/2}$  and increased with higher tempering temperature at 375°C to  $29.2 \pm 0 \text{ MPa m}^{1/2}$  and finally increased to  $92.3 \pm 1 \text{ MPa m}^{1/2}$  at 475°C. On the other hand, for ESR specimens fracture toughness improvement was only marginal, increasing to  $25.9 \pm 7 \text{ MPa m}^{1/2}$ ,  $31.0 \pm 0 \text{ MPa m}^{1/2}$  and  $92.8 \pm 1.8 \text{ MPa m}^{1/2}$  when tempered at 300°C, 375°C and 475°C, respectively.

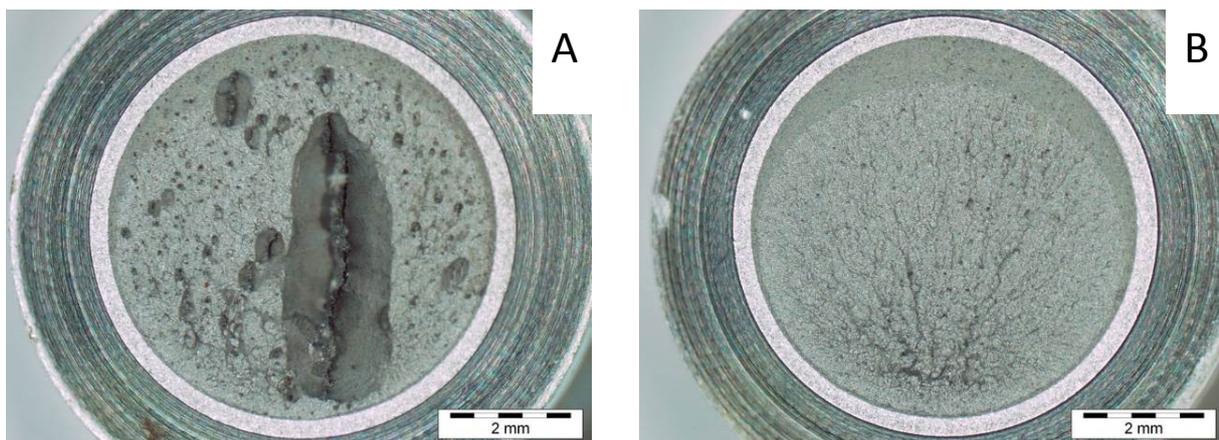


Figure 3: Fractured surface of K1c-test specimens tempered at 475°C (A) CCC and (B) ESR.

### *Fatigue resistance*

Microstructure refinement through ESR, in contrast to static mechanical properties, didn't have any positive effect on fatigue properties of the investigated spring steel. Both CCC and ESR spring steel specimens showed the same level of scattering and confidence at dynamic bending testing. Nevertheless, results of the performed tests show, that conventional continuous cast spring steel (CCC) has better fatigue resistance (Fig. 4). For CCC spring steel specimens tempered at



300°C fatigue failure at bending stress level of 330, 380 and 430 MPa occurred after approximately 120.000, 50.000 and 30.000 cycles, respectively. At the highest tempering temperature of 475°C fatigue life at given bending stress levels increased to approx. 170.000, 75.000 and 45.000 cycles. The number of bending cycles till failure at the same stress levels has been shortened for 10-15% when using ESR refinement, as clearly shown in Fig. 4.

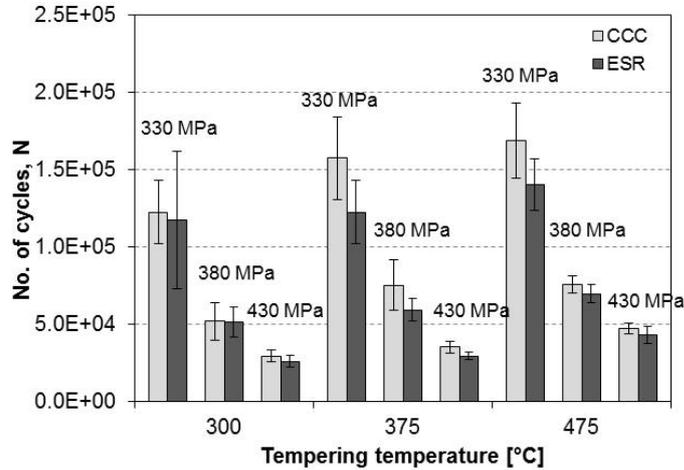


Figure 4: Effect of ESR refinement and segregations removal on bending fatigue resistance.

Effect of ESR process shown in microstructure refinement and segregations removal has even more negative effect on fatigue properties of spring steel under tensile-compression loading mode. However, these tests were carried out only for tempering temperature of 475°C. For the ESR test specimens tempered at 475°C fatigue failure at 660, 720 and 780 MPa stress level occurred after 29.000, 43.000 and 54.000 cycles, respectively. On the other hand, classical CCC specimens tempered at 475°C showed between 5 to 20% longer lifetime, lower scattering and about 5% higher tensile fatigue limit of 650 MPa, as shown in Fig. 5. These results indicate positive effect of elongated segregations being present in CCC spring steel on its fatigue resistance. However, this positive effect of segregations can be expected only when segregations are oriented perpendicular to the crack propagation direction. On the other hand, presence of some Al inclusions after ESR, as revealed by SEM microstructure analysis has further negative effect on the fatigue resistance of ESR refined spring steel, representing critical locations for crack initiation.

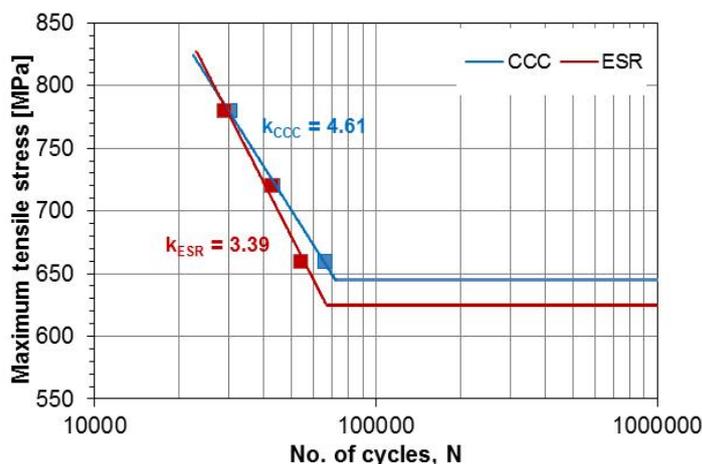


Figure 5: Tensile-compression fatigue properties of CCC and ESR spring steel, tempered at 475°C.

### Fractographic analysis

Macrographic investigation of the fractured surfaces reveals positive effect of segregations on the crack propagation resistance. It is assumed that combination of softer sulphide-type non-metallic inclusions in positive segregations (fully martensitic) and harder negative segregations (martensite and bainite) in CCC specimens retard propagation of transversal crack, thus improving spring steel fatigue resistance. Fractured surfaces of both CCC and ESR samples are shown in Fig.



6, where image A presents CCC specimen with more smoother surface in the crack propagation zone and image B presents ESR specimen with rougher and sharper fractured surface. The region of crack propagation zone is marked with black dot (Fig. 6).

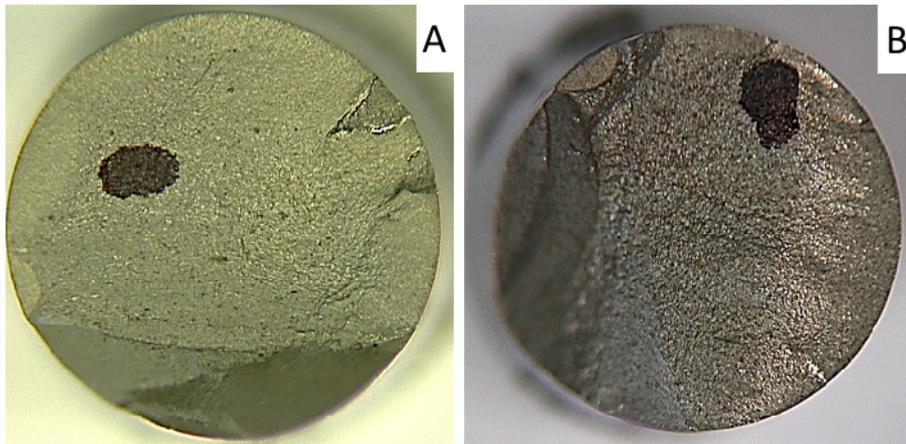


Figure 6: Macrographic image of fractured surfaces (A) CCC specimen, (B) ESR specimen.

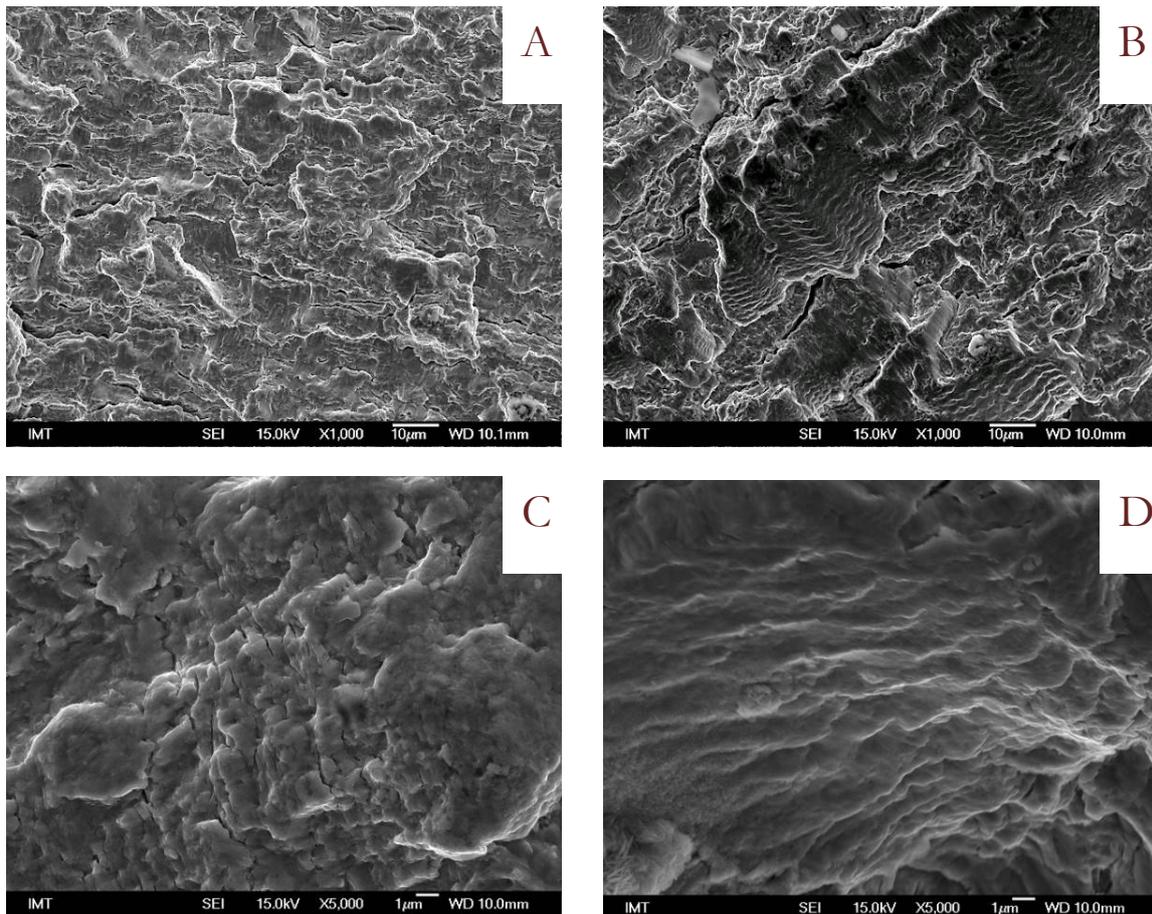


Figure 7: SEM observation of fractured surfaces (A and C) CCC specimen, (B and D) ESR specimen.



Fractured surfaces of specimens exposed to tensile-compression testing were analyzed using SEM and fatigue quasi-ductile fractured surfaces are shown in Fig. 7. The images of fractured surface are from the crack propagation zone, where obvious difference between fractured surface of CCC (Fig. 7A and 7C) and ESR (Fig. 7B and 7D) can be seen. In the case of CCC specimens fractured surface is smoother and consist of large number of separated short micro cracks being oriented perpendicular to the main crack patch through the specimen. It can be concluded that those micro cracks are formed during propagation of the main crack trough the specimen, when it reaches the softer negative segregation region in the material, containing sulphide-type non-metallic inclusions. At those inclusions the main crack changes its direction from transversal to longitudinal, thus retarding crack propagation rate. Energy needed for changes in the crack propagation direction coupled with longer crack propagation path also results in better fatigue resistance of CCC specimens. In the case of fractured surface of ESR specimens (Fig. 7B and 7D) very distinctive regions of fracture can be found, with staircase-like fracture regions being clearly visible. Fatigue test results show, that the absence of segregations, which leads to vivid larger staircase-like fracture regions and faster crack propagation result in reduced fatigue strength of the material, corresponding to reduced lifetimes of the spring steel.

## CONCLUSIONS

**A**dditional process of electro-slag remelting contributes to more uniform and refined microstructure of spring steel as well as elimination of segregations. More uniform and refined microstructure mostly results in better repeatability and smaller scattering in mechanical and fatigue properties of examined spring steel. Results of this investigation revealed that ESR process increase mechanical properties of hot rolled and heat treated spring steel. It contributes to considerable increase in impact toughness, especially at sub-zero temperatures, while increase in hardness, tensile strength and fracture toughness is less pronounced. On the other hand, ESR results in reduced fatigue resistance and fatigue limit of the spring steel investigated. For both, tensile-compressive and bending loading conditions elimination of segregations and presence of some Al non-metallic inclusions contributes to reduced cracking resistance and deteriorated fatigue properties of spring steel. In the examined fractured surfaces additional ESR process leads to formation of staircase-like fracture regions with reduced crack propagation resistance and consequently to reduced fatigue properties of spring steel. In the case of the CCC specimens crack patch is changed at softer inclusions in the positive segregations, thus leading to formation of a large number of smaller cracks perpendiculars to the main crack direction.

## ACKNOWLEDGMENTS

**A**uthors would like to thanks Štore Steel company for the financial support and to Slovenian Research agency (ARRS) for the support through research program P2-0050.

## REFERENCES

- [1] Perrard, F., Charvieux, F., Languillaume, J., A new spring steel with improved ductility dedicated for high strength parabolic leaf springs, 2nd Int. Conference Super-High Strength Steels, Peschiera del Garda, Italy, (2010).
- [2] Choi, S., Optimization of microstructure and properties of high strength spring steel, Ph.D. thesis, Posco, Korea, (2011).
- [3] Nam, W.J., Lee, C.S., Ban, D.Y., Effects of alloy additions and tempering temperature on the sag resistance of Si–Cr spring steels, *Materials Science and Engineering*, A289 (2000) 8-17.
- [4] Ona, H., Cold roll forming for high tensile strength steel sheet proposition on forming of thin spring steel sheet pipe, *Journal of Materials Processing Technology*, 153–154 (2004) 247-252.
- [5] Ai, J.H., Zhao, T.C., Gao, H.J., Hu, Y.H., Xie, X.S., Effect of controlled rolling and cooling on the microstructure and mechanical properties of 60Si2MnA spring steel, *Journal of Materials Processing Technology*, 160 (2005) 390-395.
- [6] Ardehali Barani, A., Ponge, D., Raabe, D., Refinement of grain boundary carbides in a Si–Cr spring steel by thermomechanical treatment, *Materials Science and Engineering*, A426 (2006) 194-201.
- [7] Nie, Y.-h., Hui, W.-j., Fu, W.-t., Weng, Y.-q., Effect of Boron on Delayed Fracture Resistance of Medium-Carbon High Strength Spring Steel, *Journal of Iron and Steel Research, International*, 14 (2007) 53-59.



- [8] Ardehali Barani, A., Li, F., Romano, P., Ponge, D., Raabe, D., Design of high-strength steels by microalloying and thermomechanical treatment, *Materials Science and Engineering*, A463 (2007) 138-146.
- [9] Zhang, C.-l., Liu, Y.-z., Jiang, C., Xiao, J.-f., Effects of Niobium and Vanadium on Hydrogen-Induced Delayed Fracture in High Strength Spring Steel, *Journal of Iron and Steel Research, International*, 18 (2011) 49-53.
- [10] Zhang, C.-l., Liu, Y.-z., Zhou, L.-y., Jiang, C., Secondary Hardening, Austenite Grain Coarsening and Surface Decarburization Phenomenon in Nb-Bearing Spring Steel, *Journal of Iron and Steel Research, International*, 19 (2012) 47-52.
- [11] Ganesh, P., Sundar, R., Kumar, H., Kaul, R., Ranganathan, K., Hedao, P., Tiwari, Pragya, Kukreja, L.M., Oak, S.M., Dasari, S., Raghavendra, G., Studies on laser peening of spring steel for automotive applications, *Optics and Lasers in Engineering*, 50 (2012) 678-686.
- [12] Li, W., Sakai, T., Wakita, M., Mimura, S., Effect of surface finishing and loading condition on competing failure mode of clean spring steel in very high cycle fatigue regime, *Materials Science and Engineering*, A552 (2012) 301-309.
- [13] Dziemballa, H., Manke, L., *Steel, Future for the Automotive Industry*, Verlag Stahleisen GmbH, Dusseldorf, (2005) 341-348.
- [14] Ardehali Barani, A., Ponge, D., Optimized Thermomechanical Treatment for Strong and Ductile Martensitic Steels, *Materials Science Forum*, 539-543 (2007) 4526-4531.
- [15] Assefpour-Dezfuly, M., Brownrigg, A., Parameters affecting sag resistance in spring steels, *Metallurgical and Materials Transactions A*, 20 (1989) 1951-1959.
- [16] Shin, J.-C., Lee, S., Hwa Ryu, J., Correlation of microstructure and fatigue properties of two high-strength spring steels, *International Journal of Fatigue*, 21 (1999) 571-579.
- [17] Krauss, G., McMahon, C.J., *Martensite*, ASM International, Materials Park, (1992).
- [18] Bytyqi, A., Pukšić, N., Jenko, M., Godec, M., Characterization of the inclusions in spring steel using light microscopy and scanning electron microscopy, *Materials and technology*, 45 (2011) 55-59.
- [19] Wang, X.H., Jiang, M., Chen, B., Li, H.B., Study on formation of non-metallic inclusions with lower melting temperatures in extra low oxygen special steels, *Science China Technological Sciences*, 55 (2012) 1863-1872.
- [20] Senčič, B., Leskovšek, V., Fracture toughness of vacuum heat treated spring steel 51CrV4. *Materials and Technology*, 45 (2011) 67-73.
- [21] Roberts, G., Kraus, G., Kennedy, R., *Tool Steels*. 5th edition, ASM International, Materials Park, (1998).
- [22] Tang, W., Wu, X., Min, Y., Xu, L., Effect of microstructural homogeneity on mechanical and thermal fatigue behavior of a hot-work tool steel, *Proceedings of 6th International Tooling Conference*, Karlstad University, Sweden, (2002) 755-765.
- [23] Li, W., Sakai, T., Wakita, M., Mimura, S., Influence of microstructure and surface defect on very high cycle fatigue properties of clean spring steel, *International Journal of Fatigue*, 60 (2014) 48-56.
- [24] Li, W., Sakai, T., Wakita, M., Mimura, S., Effect of surface finishing and loading condition on competing failure mode of clean spring steel in very high cycle fatigue regime, *Materials Science and Engineering: A*, 552 (2012) 301-309.
- [25] Podgornik, B., Leskovšek, V., Godec, M., Senčič, B., Microstructure refinement and its effect on properties of spring steel, *Materials Science and Engineering: A*, 599 (2014) 81-86.
- [26] Podgornik, B., Žužek, B., Leskovšek, V., Experimental Evaluation of Tool Steel Fracture Toughness Using Circumferentially Notched and Pre-cracked Tension Bar Specimen, *Materials Performance and Characterization*, 3 (2014) 1-17.
- [27] Wei, S., Tingshi, Z., Daxing, G., Dunkang, L., Poliang, L., Xiaoyun, Q., Fracture toughness measurement by cylindrical specimen with ring-shaped crack, *Eng. Fract. Mech.* 16 (1982) 69-82.
- [28] Gdoutos, E.E., *Fracture Mechanics Criteria and Applications*, Kluwer Academic Publishers, London, (1990).