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CHARACTERIZATION OF THE INCLUSIONS IN SPRING STEEL USING LIGHT MICROSCOPY AND SCANNING ELECTRON MICROSCOPY

KARAKTERIZACIJA VKLJUČKOV V VZMETNIH JEKLIH S SVETLOBNO IN VRSTIČNO ELEKTRONSKO MIKROSKOPIJO

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The quality of high-grade steel depends mainly on the size, distribution and composition of the inclusions in the steel. A good-quality product can be obtained by controlling the size, distribution and chemical composition of the inclusions. This study was limited to non-metallic inclusions, mainly compounds of metallic elements and carbon, nitrogen, oxygen or sulfur. The inclusions in spring steel were investigated with light microscopy and scanning electron microscopy. Generalized extreme value theory was applied to the size distribution of the inclusions. The types of inclusions were evaluated and classified in the majority as having a complex non-metallic composition. The experimental results show the possibility to define permissible limits for non-metallic inclusions with a minimal negative influence on the steel's properties.

Keywords: non-metallic inclusions, SEM, EDS, spring steel, GEV

Glavni dejavnik, od katerega je odvisna visoka kakovost jekla, je velikost, razporeditev in sestava nekovinskih vključkov. Z nadzorom velikosti, porazdelitve in kemijske sestave vključkov lahko dobimo kakovosten proizvod. Preiskava je bila omejena na nekovinske vključke, ki so v glavnem spojine kovinskih elementov z ogljikom, dušikom, kisikom ali žveplom. V prispevku sta karakterizacija nekovinskih vključkov in čistost vzmetnega jekla opredeljeni s preiskavami v svetlobnem mikroskopu in v vrstičnem elektronskem mikroskopu. Porazdelitev velikosti vključkov je bila opredeljena s posplošeno teorijo ekstremnih vrednosti. Iz rezultatov je bilo mogoče oceniti naravo vključkov, in sicer je bila večina vključkov kompleksnih, zraščenih z enofaznimi vključki. Rezultati eksperimentalnih postopkov kažejo na možnost določitve dopustne meje nekovinskih vključkov z minimalnim negativnim vplivom na lastnosti jekla.

Ključne besede: nekovinski vključki, SEM, EDS, vzmetno jeklo, posplošena teorija ekstremnih vrednosti

1 INTRODUCTION

Non-metallic inclusions are formed during the steel's production process and have a great impact on the strength, plasticity, fracture toughness, fatigue strength and other properties[1]. Producers can achieve better control of the processing and improve the quality of products by a characterization of the individual inclusions. The size and distribution of inclusions are particularly important, because large macro-inclusions are the most harmful for the mechanical properties of steels[2]. However, large inclusions are difficult to inspect because of their low occurrence rate. Many experimental results show that the largest inclusions are the most probable fracture origins in a given volume of material[3]. Therefore, it is important that steelmakers learn to better control the inclusions' characteristics. The characterization of non-metallic inclusions in steel in terms of number, size, shape and chemical composition is thus an essential requirement for metallurgical process technology. It is necessary to measure the size and spatial distributions of the inclusions for a correlation with the mechanical properties[4]. Different statistical models can be used to characterize the size distribution of inclusions and a lot of effort was expended in recent years to predict the limits for inclusion size or mechanical properties by extrapolating the data[5].

However, despite the major advances in inclusion control, there is still no rapid and accurate method for determining the type, size and number of inclusions present in a steel sample. The scope of possible changes in the production and characterization processes requires scientific studies.

2 EXPERIMENTAL

High-purity steel was produced in Štore Steel, d. o. o., in compliance with the EN 51CrV4 standard. The chemical composition is given in **Table 1**. Five samples with dimensions of 20 mm × 28 mm × 10 mm were cut from the slab according to the ISO 4967 standard. Samples were prepared with standard metallographic techniques, then examined by light microscopy and by scanning electron microscopy (SEM). The advantage of optical microscopy is the ability to examine a large area in a short time, but the information we are able to gather about inclusions is limited. Imaging at higher magnification and microchemical analyses were made using

SEM with an energy-dispersive x-ray spectroscopy (EDS) capability.

The samples were examined with optical microscopy in order to determine the quantity and size of the inclusions. A total of 554 images of polished samples at 100x magnification with a total area of 150 mm^2 were acquired. Image processing and measurements of the inclusion cross-section areas were made with the analySIS software.

The SEM was used in the secondary-electrons imaging and backscattered-electrons imaging modes to analyze the inclusions; details of the composition of the inclusions were investigated by EDS mapping and point analyses.

The light microscopy and SEM analyses (JEOL JSM-6500F) were performed at the Institute of Metals and Technology in Ljubljana.

Table 1: Chemical composition of the spring steel sample No. 49115 in *w*/%

Tabela 1: Kemijska sestava vzorca vzmetnega jekla št. 49115 v masnih deležih, *w*/%

Element	C	Si	Mn	Cr	V	S	Al
Composition *w*/%	0.52	0.35	0.96	0.93	0.12	0.007	0.010

3 RESULTS AND DISCUSSION

3.1 Light microscopy

Since thorough inspection is difficult and time-consuming, statistical methods have been developed for predicting the characteristic maximum inclusion size in a large volume of steel by extrapolating from data gathered on small samples. In this study, Generalized Extreme Value (GEV) theory was applied to the results and a prediction for the characteristic inclusion size was calculated.

The basic concept of extreme value theory is that when a number of data points following a basic distribution are collected on multiple samples, the maxima and minima of each of these sets also follow a certain distribution. The distribution function was given by Gumbel [6,7]:

$$G(z)=\exp\left(-\exp\frac{-(z-\lambda)}{a}\right) \tag{1}$$

where is the probability that the maximum inclusion is no larger than the size z, and a and λ are the scale and location parameters. It was first applied to the inclusion sizes in steels by Murakami and coworkers. The distribution was later generalized with the addition of another parameter ξ:

$$G(z)=\exp\left(-\left[1+\xi\left(\frac{(z-\lambda)}{a}\right)\right]^{-1/\xi}\right) \tag{2}$$

which reduces to the Gumbel distribution (Eq. 1) for ξ = 0.

A standard inspection area S_0 = 0.27 mm^2 was defined and 554 such areas were examined. The cross-section area of the largest inclusion in each inspection area was measured and a square root of the area z =# was calculated. Values of the distribution parameters were then calculated using the maximum-likelihood method. The hypothesis $\xi = 0$ was not statistically supported and thus abandoned. The parameter values of the generalized extreme-value distribution are listed in **Table 2**.

Table 2: Estimated parameters with standard errors from the GEV method for the experimental steel

Tabela 2: Ocene parametrov s standardnimi napakami za generalizirano metodo ekstremnih vrednosti

Parameter	Value	Std. error
α	1.96	0.06
λ	6.37	0.09
ξ	–0.0089	0.0018

To estimate the size of the maximum inclusion in the volume of spring steel *V*, the return level *T is* defined as follows:

$$T = V/V_0 \tag{3}$$

where $V_0 = h \cdot S_0$ is the standard inspection volume, as defined by Murakami et al., where $h = \sum \sqrt{area_{\max,i}}/N$. The estimate for the characteristic size of the maximum inclusion is then calculated from [refs]:

$$z_{\max} = \lambda - \frac{a}{\xi}\left(1-\left[-\ln\left(1-\frac{1}{T}\right)\right]^{-\xi}\right) \tag{4}$$

There exists an estimation upper limit of the inclusion size when $\xi < 0$. The characteristic sizes of the maximum inclusion in different volumes of spring steel are shown in **Figure 1**. The estimation's upper limit is 224 μm. The predicted result of the inclusion size can be used in a database for a reference in the steel-making process.

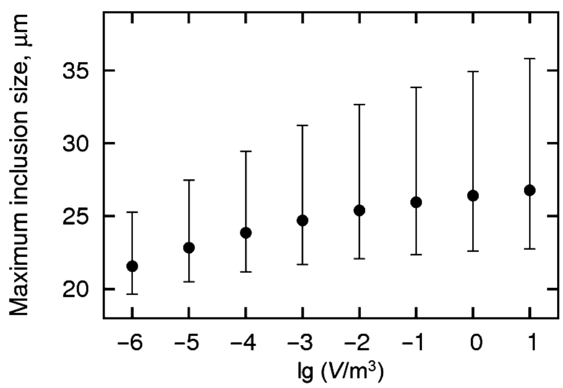

Figure 1: Estimated characteristic sizes of the maximum inclusions in different volumes. Confidence intervals were calculated using the maximum-likelihood method for a 95% confidence level.

Slika 1: Ocenjene karakteristične velikosti največjih vključkov za različne volumne jekla. Intervali zaupanja so bili izračunani po metodi največje verjetnosti za 95-odstotno stopnjo zaupanja.

3.2 SEM analysis

Scanning electron microscopy of the investigated spring-steel samples revealed different types of inclusions. **Figure 2** shows a typical complex inclusion. Three analyses were performed on this inclusion to reveal the detailed composition, mainly of calcium sulfide and aluminum oxide, **Table 2**.

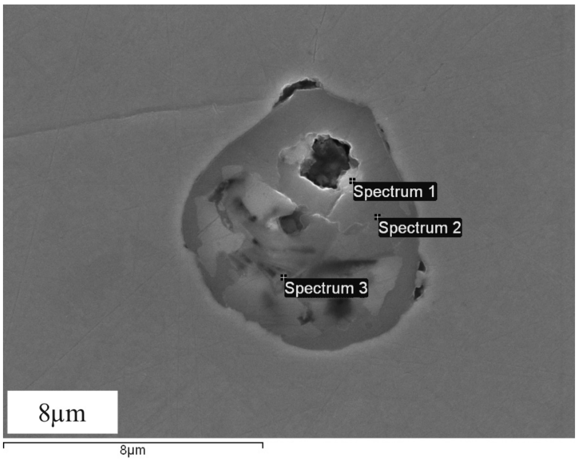

Figure 2: SEM image of a complex, non-metallic inclusion taken by secondary electrons

Slika 2: Kompleksen nekovinski vključkek, posnet s sekundarnimi elektroni v vrstičnem elektronskem mikroskopu

Aluminum oxide inclusions are the result of aluminum de-oxidation. The great advantage of steel de-oxidation with aluminum and the use of calcium to modify the form of alumina inclusions is that they reduce the dissolved oxygen activity in liquid steel, resulting in a final product with a large index of cleanliness and a smaller tendency for porosity formation[8].

Different inclusions were observed with scanning electron microscopy in sample 2. This chemical composition was similar, but the size was different. According to the EDS analysis the content of calcium sulfide (CaS) was greater than the content of aluminum oxide (Al_2O_3). As can be seen in **Figure 3**, aluminum oxide (Al_2O_3) in the inclusion center is surrounded by calcium sulfide (CaS). The distribution of the elements (**Table 3**) indicates that sulphur is bound in the manganese sulphide (MnS).

The analysis of the non-metallic inclusions in the steel 100 Cr6 indicated the presence of complex, oxide-sulfide and sulphide inclusions with different shape characteristics, while, a higher content of the sulphide inclusions is related to the high content of sulphur[9].

A complex inclusion can be seen in **Figure 4**. The detailed analysis of the inclusion supported the finding and showed that the inclusion consisted of calcium sulfide and aluminum oxide. The acquired SEM

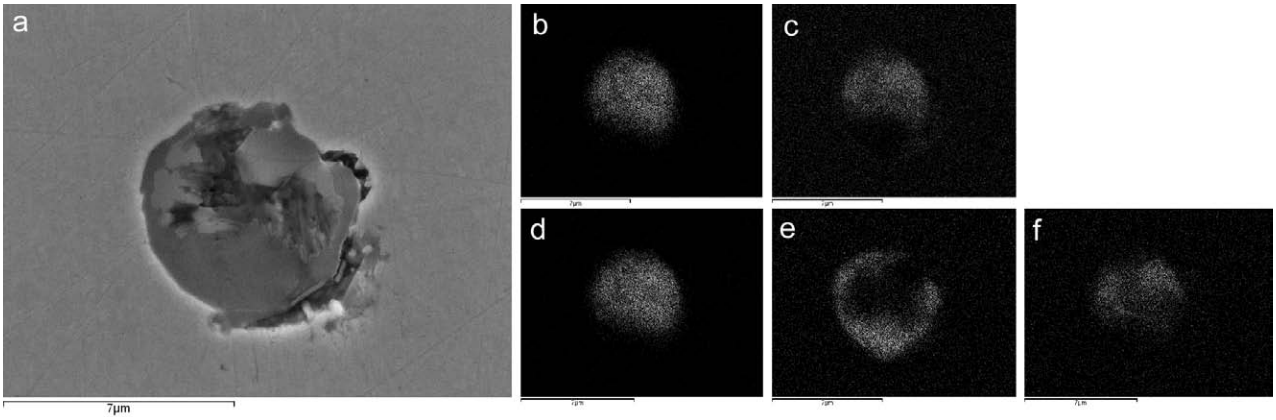

Figure 3: (a) SEM image of a complex, non-metallic inclusion and the corresponding elemental distribution for (b) O Kα1, (c) Mn Kα1, (d) Al Kα1, (e) S Kα1, (f) Ca Kα1.

Slika 3: (a) Nekovinski vključek, posnet s sekundarnimi elektroni v vrstičnem elektronskem mikroskopu in ploskovni posnetki elementov (b) O Kα1, (c) Mn Kα1, (d) Al Kα1, (e) S Kα1, (f) Ca Kα1

Table 2: Semi-quantitative chemical analysis of the inclusion in the studied spring steel in *w*/%

Tabela 2: Semikvantitativna kemijska analiza nekovinskega vključka preiskovanega vzmetnega jekla v masnih deležih, *w*/%

Elements	O	Mg	Al	Si	S	K	Ca	Mn	Fe
Spectrum 1	4.89	—	1.53	—	28.1	—	39.6	3.33	7.80
Spectrum 2	5.48	—	1.53	—	36.1	—	41.1	3.95	6.33
Spectrum 3	48.7	2.42	34.8	2.19	1.29	1.92	4.35	—	4.21

Table 3: Semi-quantitative chemical analysis of the inclusion in *w*/%

Tabela 3: Semikvantitativna kemijska analiza nekovinskega vključka v masnih deležih, *w*/%

Elements	O	Mg	Al	S	Ca	Mn	Fe
Spectrum	4.20	2.06	2.14	33.8	5.52	47.1	5.07

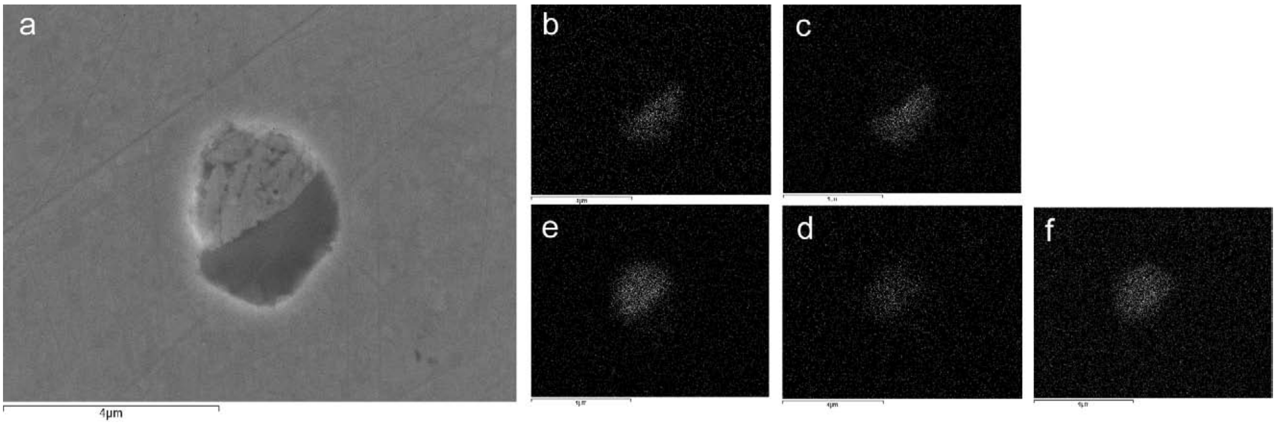

Figure 4: (a) SEM image of a complex, non-metallic inclusion and the corresponding elemental distribution for (b) S Kα_1, (c) Ca Kα_1, (d) Mg Kα_1, (e) Al Kα_1, (f) O Kα_1

Slika 4: (a) Nekovinskih vključki posneti v vrstičnem mikroskopu in ploskovni posnetki elementov (b) S Kα_1, (c) Ca Kα_1, (d) Mg Kα_1, (e) Al Kα_1, (f) O Kα_1

Table 4: Semi-quantitative chemical analysis of the inclusion in the studied spring steel in *w*/%

Tabela 4: Semi-kvantitativna kemična analiza nekovinskega vključka v masnih deležih, *w*/%

Elements	Al	S	Ca	Mn	Fe	Cr	Cu
Spectrum	0.81	26.4	0.37	48.2	22.4	0.64	—-

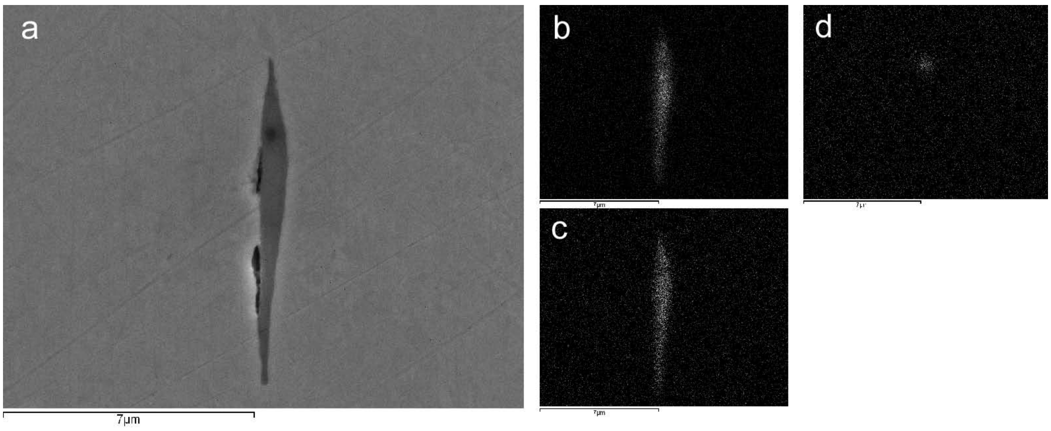

Figure 5: (a) SEM image of a complex, non-metallic inclusion and the corresponding elemental distribution for (b) S Kα_1, (c) Mn Kα_1, (d) Al Kα_1

Slika 5: (a) Nekovinski vključek, posnet s sekundarnimi elektroni v vrstičnem elektronskem mikroskopu in ploskovni posnetki elementov (b) S Kα_1, (c) Mn Kα_1, (d) Al Kα_1

Table 5: Semi-quantitative chemical analysis of the inclusion in **Figure 4** in *w*/%

Tabela 5: Semikvantitativna kemijska analiza nekovinskega vključka s **slike 4** (*w*/%)

Elements	V	S	Cr	Mn	Fe	Total
Spectrum 3	0.48	26.4	1.51	48.2	22.5	100.0

examination revealed that the sulfur and calcium content are lower in the light area of the inclusion than in the dark area. Correspondingly, the oxygen and aluminum contents are higher in the dark area compared with the light area. This indicates that the inclusion is composed of two different parts: the light part mainly consists of aluminum oxide (Al_2O_3), while the dark areas mainly consist of calcium sulfide (CaS) **Table 4**. The characteristic shape of the inclusion is globular and the size is 4 µm. It is quoted in references that inclusions with an irregular shape and sharp edges cause larger stress concentrations around the inclusions than inclusions with a smooth shape, even if there is not a significant difference in the size[10].

Manganese sulfide was found either as a singular inclusion or as an outer shell on an oxide. **Figure 5** shows an SEM image of a sulfide type of inclusion consisting of mass fractions 48.2 % of manganese and 26.4 % of sulfur **Table 5**. These inclusions are narrow and long because of their high ductility at the steel rolling temperature, and then deform and are elongated as the steel is rolled[11]. Because of their elongated shape, MnS inclusions may affect the transverse toughness of the steel[12]. Apart from the chemical composition, the size of the inclusions is crucial for the steel's properties. The results show that the critical inclusion size in the spring steel 51CrV4 was 0.14mm[13].

4 CONCLUSION

The nonmetallic inclusions in spring steel were investigated in the laboratory, and the inclusion size distribution and compositions were determined using optical microscopy according the international standard ISO 4967 and with SEM combined with EDS, respectively.

The more specific conclusions from this work are as follows. The majority of inclusions found in steel contain manganese sulfide (MnS), calcium sulfide (CaS) and aluminum oxide (Al_2O_3) or a mixture of these. A comparison between the inclusion compositions in the spring steel showed that they are very similar in terms of chemical composition. The SEV method can be effectively used to estimate the size of the maximum inclusion in different volumes of the spring steel. However, a reasonable estimation can be derived from the combined analysis of two methods, i.e., the metallographic and statistically methods. The inclusion characteristics are not sufficiently investigated for many of the commercially available spring steels and should be amended.

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