jmr&t
Journal of Materials Research and Technology
www.jmrt.com.br

ELSEVIER

abm

Original Article

Assessment of inclusion analysis via manual and automated SEM and total oxygen content of steel

CrossMark

Bruna Goulart Bartosiaki, Julio Aníbal Morales Pereira, Wagner Viana Bielefeldt*, Antônio Cezar Faria Vilela

Universidade Federal do Rio Grande do Sul, Porto Alegre, RS, Brazil

ARTICLE INFO

ABSTRACT

The search for increasingly cleaner steels has heightened the demand for additional analysis techniques, especially for the evaluation of inclusions in steel where greater cleanliness is required. A range of factors should be taken into account when selecting a particular method, in accordance with analysis objectives and in order to maximize the reliability of results. Although statistical techniques make it possible to correlate data from smaller samples with entire heats of steel, some methods are more suited to evaluating different inclusion profiles. The objective of this study was to evaluate the main characteristics of certain techniques used to study inclusions. Two of the primary methods for direct inclusion analysis of solid steel are metallographic techniques and chemical analysis, with total oxygen content used as an indirect inclusion measurement. A search of the literature identified the main advantages and disadvantages of each method, as well as the primary limitations for their use. This makes it easier to determine the most suitable methods for carrying out the desired analysis.

1. Introduction

Non-metallic inclusions cause modifications in the properties of steel (for example, in the fatigue limit); however, for some applications, the occurrence of inclusions of a certain size may result in catastrophic failures [1–5]. When non-metallic inclusions diminish the production capacity or properties of the steel in service, it is no longer considered a clean steel and requires inclusion control to ensure quality [6]. Inclusion control should specify how the inclusions are distributed in the material and the size ranges in which they are found, as well as obtain the morphologies and chemical compositions that give rise to less damaging inclusions [7]. The foremost concern is to prevent the formation of inclusions that compromise the material in question or facilitate their

---

removal when they emerge and, subsequently, to alleviate the effects using the appropriate treatment [7,8]. Understanding the behavior of inclusions during steel processing is essential in order to ensure that the final product complies with client specifications and quality standards. The most common form of detecting harmful inclusions, in line with specific quality parameters, is by characterizing them. This can be done in a number of steel processing stages [9–11], depending on the factors to be analyzed. The aim of the present study is to determine the efficiency of scanning electron microscopy and total oxygen measurements in characterizing inclusions, as well as the difficulties involved in employing these methods.

2. General aspects on the characterization of inclusions

The characterization of inclusions consists of evaluating their size, distribution, morphology and chemical composition (type of inclusion). There are a variety of techniques that can be used industrially to determine the inclusion content of steel. However, it is important to consider the size of inclusions, time needed for inspection and the volume of the material to be analyzed [12–14]. According to Hénault [12], Fig. 1 shows the detection limit in accordance with the mass of the material to be analyzed using different methods. Fig. 1 also shows that the lower the ultrasound frequency the greater the volume of material that can be analyzed. However, this increases the minimum size of the defect that the technique is capable of detecting.

Fig. 1 indicates that when the volume of the material requiring analysis increases, the minimum size of the detectable inclusions also increases. This limits the use of ultrasound methods for identifying inclusions in steel where inclusion control involves smaller inclusions, such as bearing steel. Thus, the use of smaller representative steel samples is becoming increasingly common for analysis methods that encompass smaller inclusions in their detection range. Indeed, more than one technique is needed to characterize the total inclusions population. Methods based on the surface of the sample identify inclusions with a maximum size of 20 μm, sufficient to characterize the population of micro-inclusions. Methods based on sample volumes, for inclusions larger than 20 μm, detect inclusions that can cause potentially
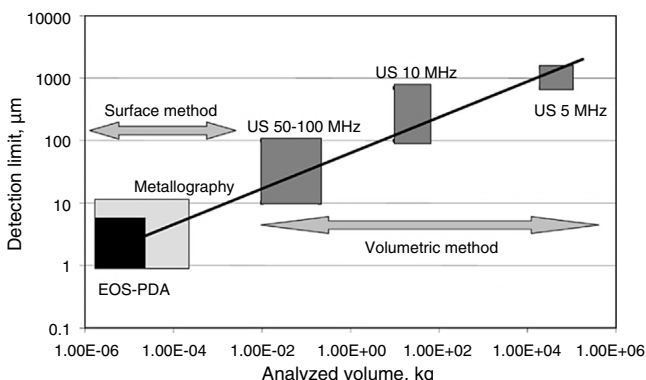
catastrophic effects during manufacturing processes or at the beginning of the life cycle of a steel part. Despite the limitation of assuming the behavior of a homogeneous heat, due to the small sample size [12], statistical methods aid in the treatment of data so that these better correspond to the reality of the entire heat [13].

According to Zhang [14], there are several methods that allow the detection of inclusions in volumes of solid steel as well as sections of solid and liquid steel. These ranges from conventional methods of like optical microscopy to ultrasound testing, optical emission spectroscopy, laser induced spectrometry, X-ray computed tomography and automated scanning electron microscopy [14,15]. For steel with low inclusion content and narrow permissible size ranges, methods with a satisfactory detection limit are necessary. Among the techniques most widely used to analyze micro-inclusions is scanning electron microscopy coupled with energy-dispersive X-ray spectrometry (SEM/EDS), which simultaneously provides an elementary chemical analysis and metallographic images. As an additional improvement, more modern equipment includes an automated system, which enables the entire sample to be scanned [16]. Indirect methods are also widely adopted in industry to determine the number of inclusions and consist of determining the chemical composition of slag, the conditions of refractory parts and even the levels of elements dissolved in the bath [14]. A commonly used technique involves measuring the total oxygen content of steel samples, which is directly related to the presence of inclusions [14,17]. Most of the equipment employed for this method simultaneously measures the dissolved nitrogen in the sample, enabling nitrogen pickup to be analyzed. The difference in dissolved nitrogen between samples from different stages of the process may be indicative of air absorption by the bath [14], which is directly related to reoxidation.

3. Sample collection for analysis

Sample type is a key variable in correctly evaluating the occurrence of inclusions and affects any analysis method adopted. Table 1 [18,19] shows the most common sample types and different characterization techniques that can be used, considering both direct and indirect methods. At times, a technique that may be used on a certain type of sample is not recommended because of difficulties associated with sampling or the analysis technique.

Immersion samplers are common in industry since they can be used both for SEM and total oxygen determination. Table 2 [20–23] shows some of the types of samplers and possible protection methods adopted.
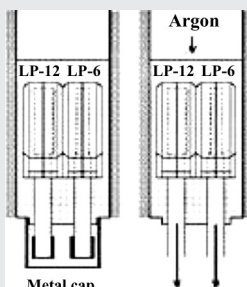
Important parameters to be considered when choosing a sampler are penetration, depth and immersion time, since these determine whether the sampler is completely filled with steel. Moreover, these parameters directly affect the cooling rate of the sample, which influences the analysis. According to Ericsson [23], the number of secondary inclusions in liquid steel samples increases significantly with the rise in the cooling rate of the samples. The protection adopted may also interfere in the results of inclusion characterization. In general, argon protection is more recommended because

Fig. 1 – Detection limit in accordance with the mass of the material to be analyzed using different methods [12].

Table 1 – Sample types and possible techniques applied [18,19].
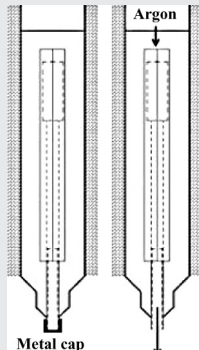
Type of sample	Analysis	Characterization techniques
Immersion samplers (steel)	Chemical composition of steel Inclusions analysis Total oxygen	Optical Emission Spectroscopy (OES), Pulse Discrimination Analysis (PDA), Oxygen Analyzer SEM/EDS
Pin (steel)	Chemical composition of steel (carbon and sulphur) Inclusions analysis Total oxygen	Optical Emission Spectroscopy (OES), Pulse Discrimination Analysis (PDA), Oxygen Analyzer SEM/EDS
Bar; Plate (steel)	Chemical composition of steel Inclusions analysis Total oxygen	Optical Emission Spectroscopy (OES), Pulse Discrimination Analysis (PDA), Oxygen Analyzer SEM/EDS
Slag	Chemical composition	X-ray Fluorescence
Clogging Mass	Chemical composition and Morphology of inclusions	SEM/EDS

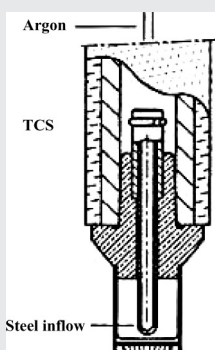**Table 2 – Immersion samplers used in industry [20–23].**

Sampler	LP-6	LP-12	BB	TOS	Samp-O-line
Thickness	~6 mm	~12 mm	~14 mm	~5 mm (Ø)	~12 mm
Metal-cap-protect	Yes	Yes	Yes	No	Yes
Argon-protect	Yes	Yes	Yes	Yes	Yes
Figure					

LP, lollipop; BB, björneborg; TOS, total oxygen sample.

samples containing deoxidizers are more easily contaminated by slag, which modifies total oxygen measurements [22]. Furthermore, deoxidizers can alter the chemical composition of inclusions in the steel sample [17]. Precautions taken with sampling depend on the factors and type of steel to be analyzed. The total oxygen sampler (TOS) prevents the entry of air and has the added advantage of minimizing sample preparation, since sectioning is not necessary [21]. However, use of the TOS is limited owing to its size, which restricts the detection of larger inclusion populations. Each type of sampler offers specific advantages and drawbacks and satisfactory results should encompass the reliability of data to be measured, compatibility with the techniques adopted and cost. Fig. 2 shows an example of using the total oxygen technique to assess cleanliness throughout the processing of CS18 steel in the furnace [5]. The vertical bars depict the mean and standard deviation for each phase of the process. Total oxygen values fall sharply along the samples, as desired.

4. Assessment of SEM and total oxygen methods: advantages and limitations

The information required in order to characterize inclusions includes their size, distribution, morphology and chemical composition. Among the possible direct methods, SEM results

meet these objectives, resulting in their widespread adoption by industry and laboratories. Nevertheless, total oxygen measurement offers the advantage of speed and simplicity in identifying inclusions in a sample already used in SEM.

4.1. Manual and automated SEM

One of the most commonly applied techniques to measure steel cleanliness is by automated SEM equipped with an EDS probe. The result is a semi-quantitative analysis of the chemical composition of inclusions on a surface prepared using conventional metallography, as well as the position, shape and size of inclusions [15]. One of the primary advantages of applying the automated method is the possibility of obtaining data for all the particles scanned in the sample [7]. Manual methods are limited to analyzing only those inclusions detected by the operator of the equipment. Moreover, there is no guarantee that the sample will be assessed equally and in its entirety. Automated SEM enables equal scanning of the sample and analyzes all the identifiable inclusions. Analysis time depends on SEM characteristics, the extent of automation, analysis parameters, sample area assessed and the cleanliness of the sample itself, among others.

The classification of inclusions in automated methods is a useful tool; however detailed knowledge of the filter
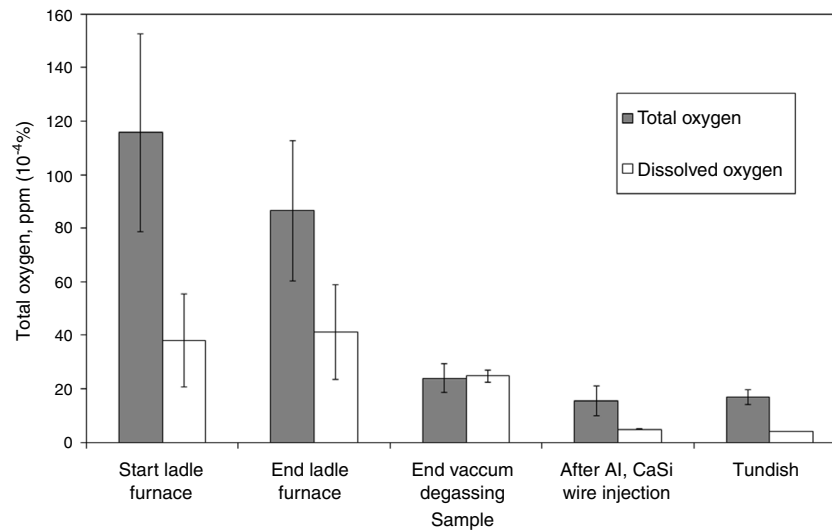
Fig. 2 – Evolution of total oxygen and dissolved oxygen for samples of CS18 steel heats [5].

system used is needed. Given that the analysis is considered semi-quantitative, results may be distorted. This is the case, for example, when it is necessary to distinguish between the different calcium aluminates formed as a result of calcium treatment in aluminum-killed steel. Table 3 shows the variation that can occur in the adjustment of SEM parameters based on different studies cited in the literature. One of the major limitations of the method is related to the representative area of the sample. Due to the combination of low inclusion occurrence in clean steel and the limited volume of the sample for cleanliness assessment, the meaning of the results is heavily dependent on the size of the area analyzed [13,25]. Another limiting factor is the size of the inclusion; in general, the larger the particle the smaller the steel matrix around the inclusion [8,11,14]. As such, quantifying the composition of micro-inclusions similar in size to the electron volume interaction (beam) is problematic and often inaccurate [28]. Most studies specify a cut-off inclusion size of 1 μm for automated analysis (Table 3).

Data obtained via automated analyses can be more reliably represented in distribution graphs, since a much larger population of inclusions can be considered in the sample. Chemical analysis of inclusions enables the chemical composition to

be depicted in ternary phase diagrams, using database management systems [7,29,30]. Limits of this analysis include normalization of the chemical composition of inclusions in ternary and pseudo-ternary systems, which may mask the real classification of the inclusion. In addition, for automated techniques, the chemical composition of scanned inclusions considers the mean of the chemical elements detected in each inclusion, therefore disregarding the occurrence of different phases in the inclusion itself, such as the agglomeration of different compounds, which is typical of exogenous [31] or oxysulfide inclusions [32]. An efficient means of characterizing inclusions is by combining manual SEM performed by an operator with automated SEM analysis. This allows the general behavior of all the inclusions to be mapped and provides a more detailed analysis of specific inclusions of interest. Another drawback of both methods is the conversion of chemical elements into non-metallic compounds, since the adoption of different criteria for the classification of inclusions produces different results. Classification criteria can be anything from simple separation between oxides and sulfides to filtering criteria for separation between different types of oxides (for example: alumina, calcium aluminates, calcium silicates, etc.). Moreover, the results obtained via manual SEM

Table 3 – Parameters adjusted in SEM.						
Author	Material	Area (mm^2)	Beam energy (kV)	Working distance (mm)	Minimum particle size (μm)	Analysis time per particle
Bartosiaki et al. [7]	SAE 52100	62	20	16–18	4	Min. 1sMax. 2s
Kaushik and Yin [18]	AHSS steel	150–180	20	–	–	–
Michelic et al. [13]	Stainless steel	100–200	15	10	4	3s
Kaushik et al. [24]	Steel	180	20	15–20	1	2s
Graham and Yang [25]	HY-100	>30	–	–	1,1	–
Verma et al. [26]	Laboratory: deoxidized Al–Mg Industry: AlKLC	–	20	–	–	–
Nuspl et al. [27]	Low carbon	–	15	18–20	1	(EDX) 3s

vary not only in accordance with the parameters of the device used and sample preparation, but also with the operator's level of experience.

4.2. Total oxygen

Total oxygen content is widely used to characterize steel cleanliness in terms of oxide inclusions and assess process improvement and quality control. Total oxygen is the sum of soluble oxygen in liquid steel and that present as oxide inclusions [7,14,17]. The major advantage of this technique is the ease and speed of obtaining values that are directly related to cleanliness and easier to correlate with meltshop process parameters, such as processing times and the chemical composition of the steel itself and slag.

The most common method for measuring total oxygen is a testing procedure using equipment that simultaneously analyzes oxygen and nitrogen in the sample [18,19]. Although this is a quick and easy technique for evaluating cleanliness during the various production stages; it is only applicable for oxide inclusions and provides no information on the chemical composition and morphology of inclusions. However, when data from this technique are used in conjunction with the results of SEM–EDS testing, for example, the final analysis is far more detailed. Industrial samples may exhibit significant variations in O_{Total} values. As mentioned in item 3, the choice of sampler and sample preparation techniques must be consistent in order for the obtained results to be compared and correlated with the process conditions of industrial heats [18]. This is a critical factor in analyzing steel with high internal cleanliness, where maximum total oxygen values do not exceed 0.0010%. The limit value is that of thermodynamic equilibrium, which is a function of the chemical composition of steel, as discussed by Holappa [33]. For aluminum-killed steels, for example, the minimum value will be between 3 and 5 ppm [33].

5. Conclusions

Conclusions reached in regard to tools for characterizing inclusions are as follows:

- Regardless of the analysis to be performed, the type of sample should always be selected as a determining criterion in obtaining representative results.
- The manner in which sampling is carried out also affects results, since penetration, depth and insertion time determine whether the sampler is correctly and completely filled with liquid steel.
- The selection of a method should take into account a number of factors, including the type of sample and the techniques used to analyze it, as well as the type of steel to be evaluated.
- Techniques that involve larger sample volumes analyzed in a viable time encompass larger inclusions in their detection limits and, as such, are not recommended for identifying micro-inclusions.
- With regard to the comparison between manual and automated SEM, automation enables data to be obtained for all the detectable inclusions in the sample scanned, ideal for

distribution analysis, for example. However, certain peculiarities, such as phases and morphologies, can be better assessed using the manual method, which analyzes each detail according to the operator's specifications.
- Total oxygen measurements can be quickly and easily obtained. Nevertheless, these measurements only consider oxide inclusions and provide no information on the inclusions present in the sample. The best way to use this analysis is combine it with other techniques.
- The main advantage of using total oxygen measurements is their efficient correlation with process data, since they represent the micro-inclusion population.
- While measurements of total oxygen scale the number of inclusions found in the sample, electron microscopy combined with chemical analysis enables the types of inclusions present to be characterized.
- Whenever possible, a combination of techniques is recommended in order to ensure the most comprehensive characterization of inclusions possible.

Conflicts of interest

The authors declare no conflicts of interest.

Acknowledgements

REFERENCES

[1] Colpaert H. In: da Costa e Silva ALV, editor. Metalografia dos produtos siderúrgicos comuns. 4ª edição atualizada por Editora Edgard Blucher; 2008.

[2] Kiessling R. Non-metallic inclusions in steel: Parts I–IV. London: The Institute of Materials; 1978.

[3] Cyril N, Fatemi A, Cryderman B. Effects of sulfur level and anisotropy of sulfide inclusions on tensile, impact, and fatigue properties of SAE 4140 steel. SAE International; 2008. p. 1–10.

[4] Pickering FB. Effect of composition and microstructure on ductility and toughness toward improved ductility toughness. Climax Molybdenum Development Co. (Japan) Ltd.; 1971. p. 9–31.

[5] Bielefeldt WV, Kohler AC, Moraes CAM, Vilela ACF, Klujszo LAC. Perfil inclusionário durante a fabricação de aços clean steel. In: 37° Seminário de Aciaria – Internacional. 2006. p. 554–63.

[6] Wünnenberg K. IISI study on clean steel. In: Le Revue de Métallurgie – CIT. 2005, October. p. 687–92.

[7] Bartosiaki BG, Reis BH, Pereira JAM, Bielefeldt WV, Vilela ACF. Characterization and study of the formation, treatment and removal of non-metallic inclusions in SAE 52100 steel. In: 19th IAS steel conference. 2013.

[8] Hollapa L, Helle ASL. Inclusion control in high-performance steels. J Mater Process Technol 1995;53:177–86.

[9] Beskow K, Jia J, Lupis CHP, Sichen DU. Chemical characteristics of inclusions formed at various stages during the ladle treatment of steel. Ironmak Steelmak 2002;29:427–35.

[10] Bielefeldt WV, Moraes CAM, Vilela ACF, Fernandes PC, Klujszo LAC. Evolução das inclusões no processamento do

aço SAE 8620 via aciaria elétrica. In: 59° Congresso Anual da ABM. 2004. p. 736–45.

[11] Bielefeldt WV, Marcon L, Moraes CAM, Vilela ACF. Uso de diferentes técnicas para caracterização de inclusões não-metálicas em aços. In: 15° Seminário de Aciaria do Instituto Argentino de Siderurgia. 2005.

[12] Hénault E. Simulation of inclusion inspection methods to obtain their statistical properties. In: Le Revue de Métallurgie – CIT. 2007, December. p. 577–84.

[13] Michelic SK, Wieser G, Bernhard C. On the representativeness of automated SEM/EDS analyses for inclusion characterization with special regard to the measured sample area. ISIJ Int 2011;51:769–75.

[14] Zhang L, Thomas BG. State of the art in evaluation and control of steel cleanliness. ISIJ Int 2003;43:271–91.

[15] Winkler W, Angeli J, Mayr M. Automated SEM–EDX cleanness analysis and its application in metallurgy. BHM 2007;152: 4–8.

[16] Story SR, Piccone TJ, Fruehan RJ. Inclusion analysis to predict casting behavior. Iron Steel Technol 2004;1:163–9.

[17] Zhang L, Thomas BG, Wang X, Cai K. Evaluation and control of steel cleanliness – review. In: 85th steelmaking conference proceedings. 2002. p. 431–52.

[18] Kaushik P, Lehmann L, Nadif M. State of the art in control of inclusions, their characterization, and future requirements. Metall Mater Trans B 2012;43:710–25.

[19] Kaushik P, Yin H. Thermodynamics, engineering and characterization of inclusions in advanced high strength steels. In: Clean steel 8. 2012.

[20] Bielefeldt WV. Estudo do Tratamento de Inclusões com Cálcio na Fabricação do Aço SAE 8620. Porto Alegre, Brazil: Escola de Engenharia, Departamento de Metalurgia, UFRGS; 2005.

[21] Tiekink WK, Maes R. Total oxygen measurements at Hoogovens Ijmuiden BOS No. 2. In: Steelmaking conference proceedings. 1994. p. 49–51.

[22] Ericsson OT. An experimental study of liquid steel sampling. Stockholm, Sweden: KTH School of Industrial Engineering and Management; 2009.

[23] Ericsson OT, Lionet M, Karasev AV, Inoue R, Jönsson G. Changes in inclusion characteristics during sampling of the steel. Iron Steelmak 2012;39:67–75.

[24] Kaushik P, Pielet H, Yin H. Inclusion characterization – tool for measurement of steel cleanliness and process control: Parts 1 and 2. Ironmak Steelmak 2009;36:561–71, 572–582.

[25] Graham S, Yang N. Representative volumes of materials based on microstructural statistics. Scr Mater 2003;48:269–74.

[26] Verma N, Pistorius PC, Fruehan RJ, Potter MS, Oltmann HG, Pretorius EB. Calcium modification of spinel inclusions in aluminum-killed steel: reaction steps. Metall Mater Trans B 2012;43:830–956.

[27] Nuspl M, Wegscheider W, Angeli J, Posch W, Mayr M. Qualitative and quantitative determination of micro-inclusions by automated SEM/EDX analysis. Anal Bioanal Chem 2004;379:640–5.

[28] Pistorius PC, Patadia A. The steel matrix affects microanalysis of CaO–Al$_2$O$_3$–CaS inclusions. In: 8th international conference on clean steel proceedings. 2012.

[29] Abraham S, Raines J, Bodnar R. Development of an inclusion characterization methodology for improving steel product cleanliness. In: AISTech conference proceedings. 2013.

[30] Fernandes M, Pires JC, Cheung N, Garcia A. Investigations of the chemical composition of nonmetallic inclusions utilizing ternary phase diagrams. Mater Character 2003;49:437–43.

[31] Cramb AW. AFS inclusions atlas – inclusion formation during foundry processing; 2013. http://neon.mems.cmu.edu/afs/afs2/window2.html [accessed 2013].

[32] Oikawa K, Ohtani H, Ishida K, Nishizawa T. The control of morphology of MnS inclusions in steel during solidification. ISIJ Int 1995;35:402–8.

[33] Holappa L. On physic-chemical and technical limits in clean steel production. Steel Res Int 2010;81:869–74.