

Influence of process parameters on hydrogen content in steel melt

Vpliv procesnih parametrov na vsebnost vodika v jekleni talini

Matjaž Knap^{1,*}, Alojz Rozman², Jakob Lamut¹

¹University of Ljubljana, Faculty of Natural Sciences and Engineering, Department of Materials and Metallurgy,

Aškerčeva cesta 12, 1000 Ljubljana, Slovenia

²Metal Ravne, d.o.o., Koroška cesta 14, 2390 Ravne na Koroškem, Slovenia

*Corresponding author. E-mail: matjaz.knap@omm.ntf.uni-lj.si

Abstract

Hydrogen content in steel melt plays an important role in determination of mechanical properties of solidified steel. In the case of too high amount of hydrogen annealing has to be applied. This process is time and money consuming.

During steel production the majority of hydrogen is removed from steel melt during the vacuum degasing process. The amount of hydrogen in steel melt after vacuuming is not only dependent on time of degasing but also other technological parameters.

Neuronal networks were used for analyses of technological parameters with influence on the hydrogen content in melt. Also their importance was evaluated. Further step towards prognostication of hydrogen content in steel melt was done with comparison between the results of predictions calculated on the basis of different databases.

As the most important factor was recognized the absolute humidity of air (AH) what is also documented in literature^[1]. It was proven that the prediction accuracy has not drastically got worse if air temperature was used instead AH. Other technological parameters have minor, but not negligible influence on hydrogen content in steel melt.

Key words: hydrogen content, steel melt and neuronal network

Izvleček

Vsebnost vodika v jekleni talini močno vpliva na mehanske lastnosti jekla v trdnem stanju. Pri preveliki vsebnosti je potrebno žarjenje, kar pomeni daljši čas in večje stroške izdelave.

Večino vodika se iz jeklene taline odstrani med vakuumskim razplinjenjem. Vsebnost vodika po razplinjenju ni odvisna samo od časa razplinjenja, ampak tudi drugih parametrov.

Vpliv tehnoloških parametrov smo analizirali in ovrednotili z uporabo nevronske mreže. Nadaljnji korak pri napovedovanju vsebnosti vodika v talini je bil narejen s primerjavo izračunov, narejenih na osnovi različnih podatkovnih baz.

Najpomembnejši vplivni faktor je absolutna vlaga zraka, kar je v skladu s podatki v literaturi^[1]. Ugotovili smo, da zamenjava najpomembnejšega parametra z drugim, temperaturo zraka, drastično ne poslabša napovedi. Drugi tehnološki parametri imajo manjši, ampak ne zanemarljiv vpliv na vsebnost vodika v jekleni talini.

Ključne besede: vsebnost vodika, jeklena talina in nevronske mreže

Introduction

Hydrogen in steel can cause a lot of defects what is very well documented in the literature. Over 40 articles can be finding in Science Direct with key words “steel”, “hydrogen” and “defect” solely for the year 2013. Some defects caused by hydrogen content in steel^[2,3] can be repaired with thermal treatment, which is particularly in the case of large ingots very time consuming and also uneconomic. Better way to deal with the problem of too high hydrogen content in steel melt is to reduce it during the process of secondary steel making and with appropriate casting technology.

To control the amount of hydrogen during steelmaking it is necessary to identify the parameters, which have the main influence. Several studies were conducted worldwide in last few years, which give some insight in the problematic, i.e. from S. Misra^[1], H. E. Hurst^[4] and R. J. Fruehan^[5].

Also in Slovenian steel-works the risk of too high hydrogen content in steel melt exists. The most seriously is this problem indicated at casting of large ingots. This problematic was presented in works of the researchers from Metal Ravne.^[6,7] In the past few years' studies with the intention to identify the influential parameters and consequently to lower the hydrogen amount in the steel melt were made also in collaboration between Department of Materials and Metallurgy and Slovenian steel-works.^[8-10]

Mentioned studies have identified only the influence of separate parameters. Our experiences with neuronal networks, which were successfully employed in case of characterisation and prediction of technological parameters^[11-13], have encourage us to find the correlation between them.

Experimental work

For the analyses and predictions program Statistica Neuronal Networks was used which enables use of various types neuronal networks and this makes it suitable for solving problems from different areas, for regression as well as classification cases. In the case of hydrogen content prediction – regression problem, MLP

(multilayer perceptrons) type neuronal network was used. The schematic representation of this neuronal network with three layers is shown at Figure 1.

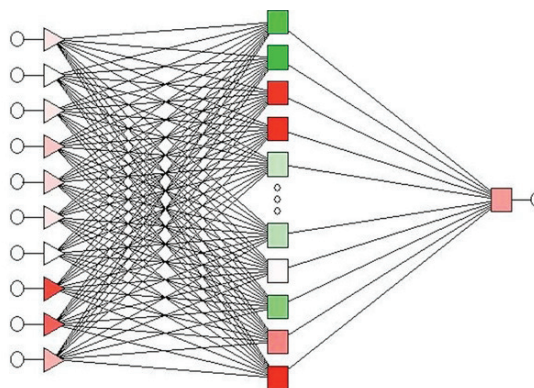


Figure 1: MLP neuronal network with ten input parameters in first layer, fifteen neurons in hidden layer and one output parameter.^[11]

For usage of neuronal nets the reliable database has to be collected and prepared for further work. Some measurements from the working process are automatically saved into the centralised database therefore the majority of necessary data was already in the system.

In the case of industrial measurements obtaining the representative database is always the important step. If data base is incomplete, unreliable or even wrong, parameters, which do not have any correlation with hydrogen content in steel melt could be regarded as crucial. Measurements and construction of initial data base was carried-out at Metal Ravne.

From the bulk database, which was composed from automatically included measured data as well as manually inserted records the uniform table was made. This table was built-up from 14 parameters (2 outputs and 12 inputs) and more than 2 500 records, i.e. data vectors.

The hydrogen content in steel melt after vacuum treatment was measured with two methods, with direct measurements with Hydris device^[4] and indirect with chemical analysis from solid sample.^[8] Hydris measurements were estimated as more reliable and thus used in this study. This decision has decreased number parameters to 13 and number of records to 1 771. Another step was filtration of the database. The filtration process was made with the help of basic statistic tools that has enabled identification

of data with unreliable extreme values. In spite of the fact that neuronal networks can very good compensate some inconstancies in data the false data on the boundary of the analysed area can cause unrealistic predictions. This filtering process also did not have big influence on the number of available data, only a little more than 1 % of records were excluded.

From the remaining data table with 1 749 and 13 parameters were built. As target parameter – output data, hydrogen content measured with Hydrys device was selected.

It is clear that steel grade could have some influence on the hydrogen content in the steel melt but it can be indirectly described with other parameters from data base.

After these reductions final database was formed. It consists of 1 749 records, so-called data vectors, with 12 parameters (11 inputs and 1 output). Database was then randomly divided into training, test and verification databases in ratio 75 : 15 : 10 what is commonly practice in analysis with neuronal networks.

First neuronal networks were used on the whole database and analysis of impact of parameters was made (Table 1). From this table it can be clearly seen that most influential parameter is absolute humidity of air (AH) what is in accordance with the results described in work of Misra.^[1]

Because of very scattered data in training database what can be clearly seen at Figure 2 the very accurate predictions are not possible, i.e. generalisation is expected. With this in mind the predictions of hydrogen content in steel melt were first made only with one input parameter – absolute humidity of air. Regarding to Table 1 also temperature (T), time of vacuum treatment (t_{vacuum}) and mass of lime (m_{CaO}) have certain influence on hydrogen content in steel melt and thus better predictions can be expected if they were included. These parameters were in fact taken into account in different combinations. For the final estimation of the

maximum accuracy of the hydrogen content description all 11 input parameters were used in neuronal network analysis.

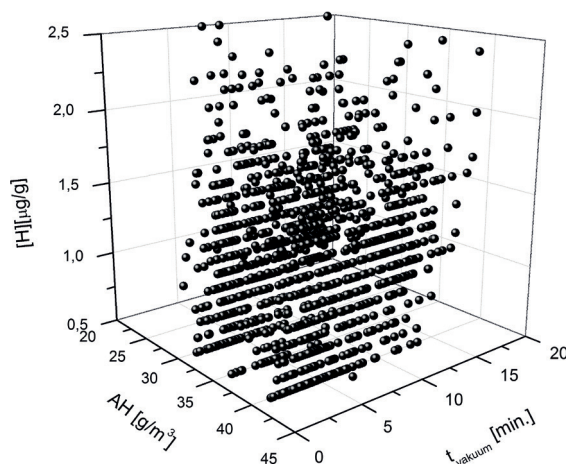


Figure 2: Influence of absolute humidity of air and time of degassing on the hydrogen content in steel melt.

Results and discussion

The successfulness of prediction is commonly defined with the least squares method (R^2) and this measure was also applied in this study. Comparison between predictions with different neuronal nets trained with various numbers of input parameters is presented in following subsections.

Influence of restricted database on the hydrogen content

When industrial data is used as input parameter for predictions it is possible that some of the regularly used parameters are not available. To find out if replacements of normally used input parameters are possible and how they affect the accuracy of predictions trials with different number of input parameters were made.

Table 1: Sensitivity analysis of influential parameters

m (kg)	t_{process} (min)	t_{vacuum} (min)	T (°C)	m_{CaC_2} (kg)	m_{boksit} (kg)	m_{Al} (kg)	m_{ferro} (kg)	m_{carbur} (kg)	m_{CaO} (kg)	AH (g/m³)
1.004 2	1.005 3	1.042 5	1.067 0	1.001 9	1.004 3	1.012 4	1.000 8	1.016 8	1.050 2	1.136 8

– *Absolute humidity of air*

In this case only absolute humidity of air was used as input parameter. From the results presented in Table 1 is clear that this parameter has the most important role in the prediction of hydrogen content. Despite quite big scattering of measured data the R^2 for these predictions was 0.433.

– *Absolute humidity of air and air temperature*

From the results of neuronal network training collected in Table 1 the second most influential parameter is air temperature. Air temperature and absolute humidity of air are not entirely independent what can be seen from Figure 3 and that is why the correlation between measured and predicted values has not drastically changed; R^2 for these predictions was 0.440.

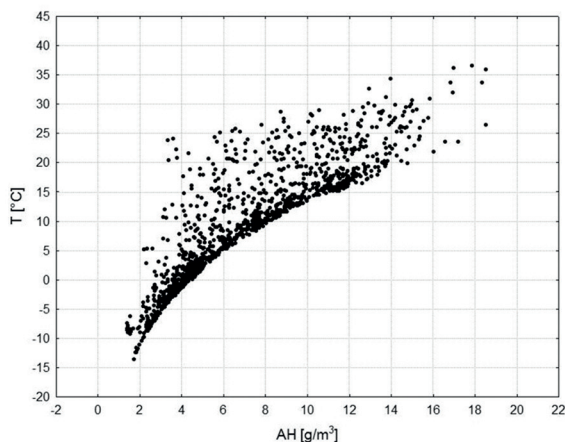


Figure 3: Relationship between absolute humidity of air and temperature.

– *Absolute humidity of air and lime mass*

With only two input parameters, absolute humidity of air and mass of added lime, the correlation between the measured and with neuronal network predicted data was significant better than with only one parameter – absolute humidity of air. The correlation factor was 0.488.

– *Absolute humidity of air, lime mass and time under low pressure (vacuum)*

From the logical point of view it is clear that the time in which the melt is under low pres-

sure must have an influence on the hydrogen content. The results of the predictions with neuronal nets have confirmed that with improved accuracy and correlation factor rises to value 0.513.

– *Predictions without absolute humidity of air as input parameter*

Two different predictions were made. In first predictions temperature of air and mass of lime were used as input parameters. The correlation factor was 0.451 what is slightly better than result when only absolute humidity of air was used as input parameter and at the same time less accurate as combination of absolute humidity of air and lime mass.

For second predictions mass of lime and time of steel melt under low pressure were used as input. The correlation factor 0.222 suggests that those two parameters don't have major influence on the hydrogen content in the steel melt.

Influence of all eleven input parameters on the hydrogen content

It is quite common that for the prediction all available data is used. The correlation factor was 0.553 what is understandingly better than at all previously mentioned efforts. Because of the scattered input data and thus quite big generalisation of predictions the risk of overtraining is not present. This can be also deduced from comparison of correlation factors for train, test and verification database. The results with weighted averaged correlation factor are presented in Table 2.

Table 2: The least squares values for three databases with 11 input parameters

Train	Test	Verification	Average
0.555	0.534	0.515	0.548

Hydrogen content as function of various parameters

The influence of absolute humidity of air was found as predominant. In this analysis further three parameters were studied: temperature of the air, the mass of lime added for slag forma-

tion and time of low pressure. The influence of these three parameters together with the main parameter are presented and discussed in further paragraphs.

— Influence of air temperature

The hydrogen content in steel melt is bigger in the case of higher absolute humidity of air. The lowest limit of absolute humidity of air increases with the air temperature growth as can be seen from Figure 3. That is why increase of hydrogen content is in steel melt with rising air temperature expected and logical. The graphical interpretation is presented at Figure 4.

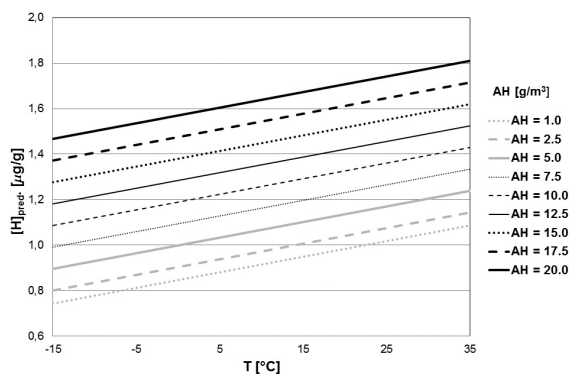


Figure 4: The influence of air temperature and absolute humidity of air on the hydrogen content.

— Influence of added mass of lime

Lime usually contains some amount of moisture. At elevated temperatures and in the presence of carbon hydrogen can be produced from steam^[14], consequently the amount of hydrogen in melt increases. The rise in the hydrogen content in the steel melt with the larger amount of added lime is clearly noticeable at the Figure 5.

From the diagram it can be deduced that the gradient of hydrogen content increase is bigger at smaller values of added lime mass. Also the influence of absolute humidity of air is visible. In the case of lower amounts of absolute humidity of air the increase of hydrogen content is evident at whole region. On the other hand at high values of absolute humidity of air increase of hydrogen content in melt is restricted only on the difference between addition and no addition.

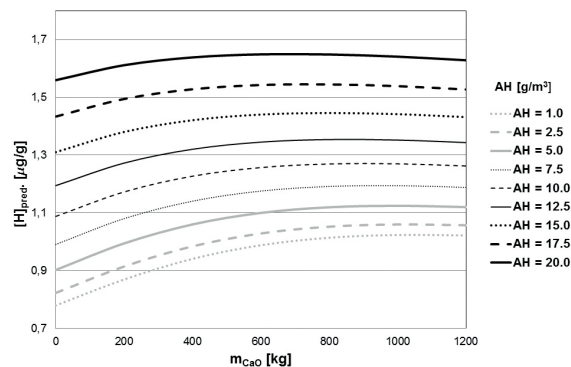


Figure 5: The influence of added mass of lime and absolute humidity of air on the hydrogen content.

— Influence of time under low pressure

The longer times of vacuuming logically leads to the lower values of hydrogen content in the melt. At the diagram at Figure 6 this assumption is confirmed.

The analysis also shows one unexpected result. It was presumed that with longer times the change in the hydrogen content will become slower but the results does not confirm that assumption.

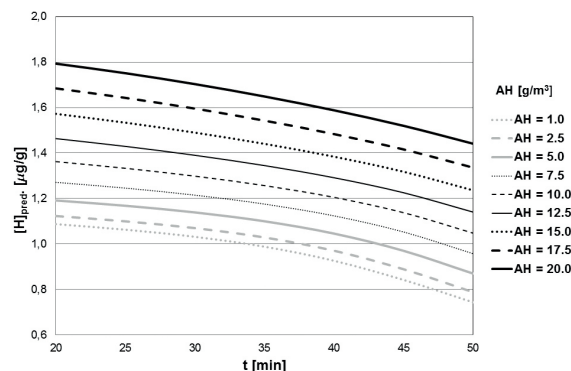


Figure 6: The influence of time of degassing and absolute humidity of air on the hydrogen content.

Comparison with the references

The hydrogen content in our study was measured after the degassing process unlike the most of data published in literature. In spite of that the conclusions from this study are in agreement with the literature. The influence of absolute humidity of air is predominant in this study and also in study from Misra.^[1] Also the rise of hydrogen content in the steel mold with increased amount of lime is in agreement with results published by Fruehan.^[5]

Conclusions

The very good correlation between the measured hydrogen content in steel melt and results of neuronal networks prediction was not expected. The main reason is very big scatter in measured results. The predictions were thus more global and orientated into the estimation of loose rules which can help to predict the tendencies of hydrogen amount change during the process of steelmaking.

Some conclusions from the analysis:

- Absolute humidity of air is the parameter which has the most important influence on the hydrogen content in the steel melt. The drastic change of other influential parameters can compensate only smaller changes in absolute humidity of air.
- Temperature is in correlation with the absolute humidity of air and thus can be alternatively used as input parameter in the case of lack of data for absolute humidity of air.
- Despite the fact that other measured parameters, e.g. mass of added lime, time under low pressure, have minor influence the prediction accuracy increases. On the other hand with only this data as input parameter the prediction of hydrogen content are not possible.
- At higher air temperatures higher values of hydrogen content in the melt are expected.
- Addition of lime during steelmaking also increases the amount of hydrogen in the melt. The rise is more noticeable at lower masses and lower absolute humidity of air.
- Longer times of vacuuming (melt under low pressure) leading to the lowering hydrogen content in the steel melt.

References

- [1] Misra, S., Li, Y., Sohn, I. (2009): Hydrogen and Nitrogen Control in Steelmaking at US Steel. *Iron and Steel Technology*, pp. 43–52.
- [2] Doshida, T., Nakamura, M., Saito, H., Sawada, T., Takai, K. (2013): Hydrogen-enhanced lattice defect formation and hydrogen embrittlement of cyclically prestressed tempered martensitic steel. *Acta Materialia*, 61, pp. 7755–7766.
- [3] Murakami, Y., Kanezaki, T., Sofronis, P. (2013): Hydrogen embrittlement of high strength steels: Determination of the threshold stress intensity for small cracks nucleating at nonmetallic inclusions. *Engineering Fracture Mechanics*, 97, pp. 227–243.
- [4] Hurst, C., Vergauwens, I. M. (2004): Gas Analysis in Steel: Identifying, Quantifying, and Managing Hydrogen Pick Up in Steel. *Annual Australian Foundry Institute Conference*, pp. 185–193.
- [5] Fruehan, R. J., Misra, S. (2005): *Hydrogen and Nitrogen Control in Ladle and Casting Operations*. Pittsburgh, PA p. 54.
- [6] Buhvald, A., Rozman, A. (2011): Super čista jekla - prihodnost za podjetje Metal Ravne. 15. Seminar o procesni metalurgiji jekla. University of Ljubljana, pp. 9–17.
- [7] Rozman, A. (2012): Kontrola vsebnosti vodika v jeklu in njegovega vpliva na nastanek kosmičev. *SIJ*, 7–8, pp. 23–24.
- [8] Vrbek, K. (2013): *Measuring hydrogen content during steelmaking*. Diploma work. Ljubljana: University of Ljubljana, p. 71.
- [9] Šuler, B. (2013): *Hydrogen in tool steels*. Diploma work. Ljubljana: University of Ljubljana, p. 44.
- [10] Turščak, J. (2011): *Influence of proces parameters on alloyed steel melt dehydration*. Diploma work. Ljubljana: University of Ljubljana, p. 65.
- [11] Knap, M., Falkus, J., Rozman, A., Lamut, J. (2008): The prediction of hardenability using neuronal networks. *Archives of metallurgy and materials*, 53, pp. 761–766.
- [12] Večko Pirtovšek, T., Kugler, G., Godec, M., Terčelj, M. (2012): Three important points that relate to improving the hot workability of ledeburitic tool steels. *Metallurgical and materials transactions. A, Physical metallurgy and materials science*, 43, pp. 3797–3808.
- [13] Večko Pirtovšek, T., Peruš, I., Kugler, G., Terčelj, M. (2009): Towards improved reliability of the analysis of factors influencing the properties on steel in industrial practice. *ISIJ international*, 49, pp. 395–401.
- [14] Chen, W.-H., Lin, M.-R., Yu, A.B., Du, S.-W., Leu, T.-S. (2012): Hydrogen production from steam reforming of coke oven gas and its utility for indirect reduction of iron oxides in blast furnace. *International Journal of Hydrogen Energy*, 37, pp. 1748–1758.