DUCTILE-TO-BRITTLE IMPACT TRANSITION TEMPERATURE FOR LOW-CARBON MICROALLOYED STEELS WITH HIGH NIOBIUM CONTENTS. A STATISTICAL APPROACH

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ABSTRACT

In the present investigation, the effect of both: rolling parameters (2 reduction rates and 3 cooling rates) and chemical elements such as: C, Mn, Nb, Ti, Mo, Ni, Cr, Cu and B, has been studied in relation with toughness properties in low-carbon microalloyed steels with high niobium contents (up to 0.12 wt.% Nb). For this purpose, an experimental set-up was designed based on an intelligent design of experiments (DoE), resulting in 26 casts (laboratory casts). A combination of metallography, Electron Back-Scattered Diffraction (EBSD) and Charpy impact tests have been performed to study how processing parameters and chemical composition affect toughness, and to generate microstructure-toughness relationships. The results, where *ITT* 27J and 0.5Kvmax are the response variables, have been analysed statistically by means of multiple linear regression technique, leading to response equations. From the results, it was found that high niobium additions improve the toughness; where its effect might be related mostly to grain size refinement.

KEY WORDS: microalloyed steels, toughness properties.

1. INTRODUCTION

Strength and toughness are two of the most important mechanical properties for the design of steel structures, pressure vessels, pipelines or other similar components [1]. Thermo-mechanical rolling is used to maximise grain refinement and thus achieve both higher strength and toughness [2]. A fine grain microstructure is an optimum method for improving strength since unlike most other strengthening mechanisms, the improvement in strength is also accompanied by an improvement in toughness The use of niobium in low-carbon bainitic steels is advantageous because when the amount of solute niobium is increased, retardation of austenite recrystallization is observed at significant higher temperatures, and also because of its ability to promote the formation of bainite [3,4].

2. INTELLIGENT DESIGN OF EXPERIMENTS

To study the effect of C, Mn, Nb, Ti, Mo, Ni, Cr, Cu and B, a statistical approach is used, by means of an intelligent design of experiments using a three-stage approach:

- ☑ In stage 1, a half fractional factorial design is used to examine five factors at two levels using sixteen casts, the factors being Mn, Ni, Cu, Mo and Cr; 16 casts in total.
- ☑ Stage 2, with combinations of low and high levels of C and Nb, was designed to check the limit conditions of High Temperature Processing (HTP) concept. The design is full factorial.
- ☑ Stage 3 investigates the influence of B and Ti. Since only B in solid solution is effective for the phase transformation

The levels for each element are shown in Table 1.

Table 1. Intelligent Design of Experiments, Laboratory casts

Stage	Level	C	Nb	Ti	B	Mn	Ni	Mo	Cu	Cr
1	Low					1.5	0	0	0	0
	Base	0.04	0.10	0.015	0					
	High					2.1	0.5	0.3	0.5	0.5
2	Low	0.01	0.04							
	Base			0.015	0	1.8	0.25	0.15	0.25	0.25
	High	0.07	0.07							
3	Low			0.008	0.000					
	Base					1.8	0.25	0.15	0.25	0.25
	High			0.025	0.002					

In total, 26 casts: 24 casts with the aim composition from the experimental design plus 2 failed casts (with high carbon level) have been made and from each cast one 12 mm thickness plate have been rolled under six conditions. These conditions have reduction ratios below the no recrystallization temperature (T_{nr}) of 2 and 4, with a finish rolling temperature of 850°C and cooling rates between 850°C and 550°C of 0.5°C/s (Air Cooling, **AC**), 10°C/s followed by air cooling (ACcelerated Cooling + Air Cooling, **ACC + AC**), and 10°C/s followed by slow cooling (ACcelerated Cooling + Coiling simulation, **ACC + CT**). All these plates have been supplied by OCAS ArcelorMittal R&D (Belgium).

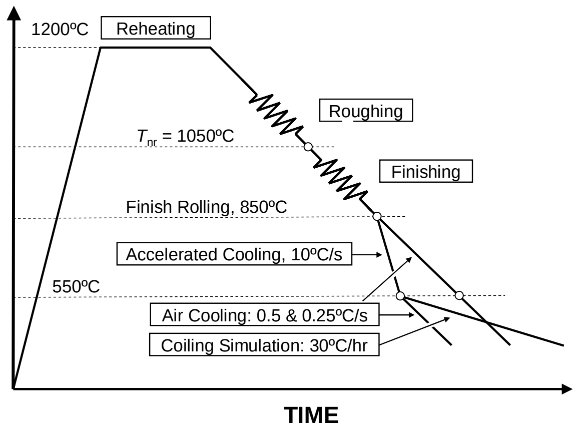


Figure 1. Schematic illustration of the thermomechanical rolling schedule of laboratory casts

Table 2 summarizes the six rolling conditions. These conditions are selected to simulate the slow cooling conditions in heavy plate mills without accelerated cooling, faster cooling in heavy mills with accelerated cooling, and in hot strip mills with coiling after rolling.

Table 2. Rolling Schedules

Rolling Schedules	Cooling Conditions		
Reduction Ratio (RR)	AC	ACC + AC	ACC + CT
$RR = 2$	C	E	G
$RR = 4$	D	F	H

Four industrially casts were used for the verification/validation of the models obtained with the laboratory casts. These industrial casts were delivered by Ruukki, Corus and Salzgitter,

Table 3. Analysis (wt. %) of the industrially cast materials

Cast	C	Mn	N	Ti	Nb	V	Ni	Mo	Cu	Cr	Ti/N
81351	0.042	1.97	0.0084	0.015	0.10	0.013	0.21	0.005	0.21	0.98	1.79
02098	0.079	1.66	0.0050	0.003	0.04	0.079	0.06	0.004	0.06	0.04	0.58
16685	0.047	1.73	0.0082	0.018	0.10	0.009	0.04	0.069	0.04	0.27	2.20
81913	0.05	1.58	0.0059	0.016	0.10	0.007	0.16	0.005	0.25	0.26	2.71

3. EXPERIMENTAL METHODS

3.1. EBSD Technique

The samples for Electron Back-Scattered Diffraction (EBSD) observations were prepared from rolling schedules samples, taking into account the rolling direction. All the scans were carried out on a Philips XL30cp Scanning Electron Microscope (SEM) at the quarter plate thickness position.

The EBSD specimens were tilted 75º from the horizontal, so that the surface was normal to the electron beam. The step size was 0.4 microns and scan size was 160 × 100 microns. TSL OIM Analysis 4.6 software was used to analyse the data.

In order to quantify the final grain size (bainitic packets in the case of bainitic microstructures) in relation with toughness, the grain size was determined with a threshold misorientation of 15º. It is widely known that the Impact Transition temperature (*ITT*, ºC) decreases as the grain size is refined; there is a strong effect of grain refinement on the brittle fracture stress [5]. The high angle boundaries show a resistance to brittle cleavage fracture, hindering the propagation of a cleavage crack.

3.2. Charpy Impact Tests

Charpy impact tests were performed by Aachen University (RWTH, Germany). Such tests were carried out to EN 10045 using an impact testing machine with an energy capacity of 1448 J and an impact velocity of 7.74 m/s. Impact transition curves were determined by means of the modified tanh fitting algorithm of Wallin [6]. Impact transition temperatures (*ITT*) were calculated following two criteria; the first one was for 27J and the second one was for half the upper shelf energy, described as 0.5Kvmax, leading to two response variables.

4. STATISTICAL ANALYSIS

4.1. Introduction

Multiple linear regression technique was used to obtain response equations from the response variables (ITT 27J and 0.5Kvmax). *Essential Regression and Experimental Design of Chemist and Engineers software*® was employed for this purpose. The sample was formed by the aforementioned 26 casts (laboratory casts). Only those parameters/variables considered in the three intelligent designs were taken into account: Chemical elements (**C, Nb, Ti, Mn, B, Cr, Cu, Ni and Mo**) and rolling parameters: **rolling reduction (*RR*)** and cooling rates (where $CR_{800\text{-}550^{o}C}$ and $CR_{550\text{-}20^{o}C}$ denote $\boldsymbol{CR_1}$ and $\boldsymbol{CR_2}$, respectively). Additional models were obtained introducing the grain size ($D_{15^{o}}$) as regressor in the response equation for toughness.

A transformation of variables takes place concerning the cooling rates (CR_1, CR_2). Note that the cooling rates are basically between 0.5 - 10ºC/s and 0.25 - 0.008ºC/s for CR_1 and CR_2, respectively. The cooling rates variables differ by orders of magnitude, being such orders of magnitude which make the difference. Therefore the cooling rates are introduced as the decimal logarithms, $\log_{10}CR_1$ and $\log_{10}CR_2$, but for the sake of brevity such terms will be spelt as $\log CR_1$ and $\log CR_2$ from now on.

4.2. Coefficients of multiple determination

In order to figure out whether a model actually describes the data adequately or how good is the “fit” of the predicted data compared to the "real data", the most common coefficient used is the **coefficient of determination, R^2**. An R^2 of 1.0 indicates that the regression line perfectly fits the data. However a R^2 value close to unity does not necessarily guarantee a good model; each additional variable added to the model increases R^2. Thus, R^2 can be made larger simply by adding more predictor variables to the model.

There is another coefficient of determination which bears in mind the degrees of freedom in the model: **adjusted coefficient of determination, $R^2_{adjusted}$**. This adjusted R^2 does not automatically increase when new predictor variables are added to the model, in fact, the R^2_{adj} may actually decrease. This gives an idea of how much o how little added value is obtained from a bigger model. Finally, another important parameter in the data analysis is the concept of **significance**, denoted as α. In statistics, a coefficient is significative if it is unlikely that occurs by chance. *The smaller its significance parameter, the safer the coefficient is.* Usually, significance levels of 0.10 and 0.05 are used to determinate whether a coefficient is significative or not. In this work, a significance level of **α = 0.1** has been adopted.

4.3. Response equations

For each parameter, a total of three equations are fitted:

- ☑ ***"Autofit"***: Only those variables with significance α **< 0.1** are considered.
- ☑ ***"R^2_{adj}"*** where all possible models are considered and the one with the largest adjusted determination **(maximum R^2_{adj})** coefficient is retained, and
- ☑ ***"Regress"*** This method considers all the variables, irrespective of their significances. This method obtains the largest determination coefficient **(maximum R^2)**.

The first option (Autofit) is the safest statistical approach. The second one is trying to detect some more effects, but the coefficients are not so sure (α > 0.1). The third option (regression to all variables) should be used with a lot of precaution: It is the best fit to the experimental set of 26 considered casts, but one has to beware about the uncertainty/significance of some coefficients (α >> 0.1).

5. RESULTS AND DISCUSSION

Two groups of models have been generated for the response variables *ITT* 27J and 0.5Kvmax, as shown in Table 4. The only difference between both is that the grain size is introduced as $D_{15^\circ}^{-1/2}$ ($mm^{-1/2}$), like Hall-Petch equation in the second group (Table 4.b). Those coefficients with significances α < 0.1 (credible ones) are shown in bold. From each response equations, 3 statistical parameters are shown, coefficient of determination (R^2), adjusted determination coefficient (R^2_{adj}) and the maximum significance (α) which presents one of the predictor variables (underlined).

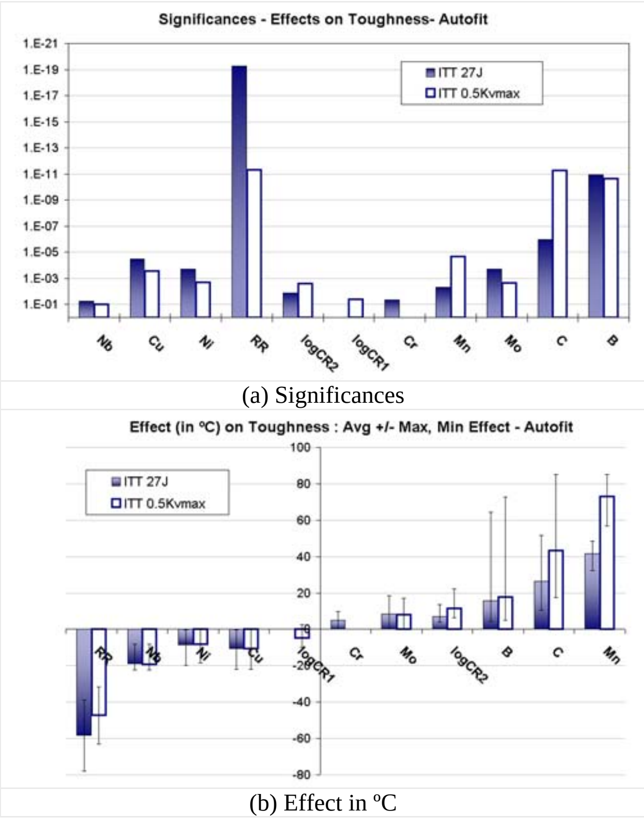
Table 4. Response equations of Toughness models: without considering the grain size (a), and considering it (b)

(a)

Ductile to brittle transition temperatures Models (ºC, weight %, log(K/s))	R^2	R^2_{adj}	α_{max}
ITT 27J = -125+22000B+550C -180Nb+55Mo-40Cu-40Ni+25Mn+20Cr-20RR -7logCR₂	0.632	**0.607**	**0.059**
= -125+22000B+550C -180Nb+55Mo-40Cu-40Ni+25Mn+20Cr-20RR -7logCR₂	0.632	**0.607**	**0.059**
= -125+22000B+550C+400Ti-200Nb+55Mo-40Cu-40Ni+25Mn+20Cr-20RR -8logCR₂-2logCR₁	**0.635**	0.604	0.515
0.5Kvmax = -150+24000B+925C -180Nb+50Mo-40Cu+40Mn-35Ni -15RR-11logCR₂-8logCR₁	0.597	**0.569**	**0.094**
= -150+24000B+925C -180Nb+50Mo-40Cu+40Mn-35Ni -15RR-11logCR₂-8logCR₁	0.597	**0.569**	**0.094**
= -150+24000B+900C-600Ti-160Nb+45Mo-40Cu+40Mn-35Ni+12Cr-15RR-11logCR₂-8logCR₁	**0.602**	0.568	0.378

(b)

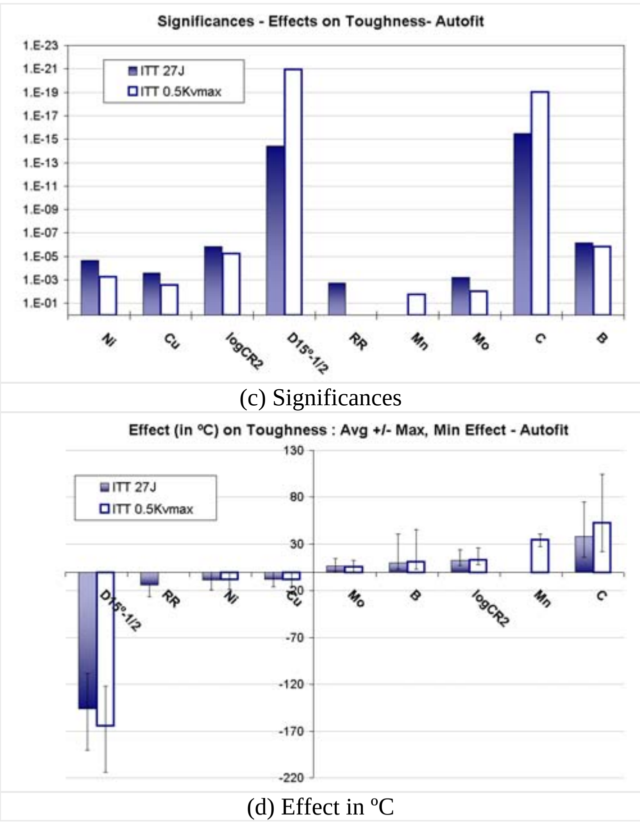
Ductile to brittle transition temperatures Models (°C, weight %, log(K/s), $mm^{-1/2}$)	R^2	R^2_{adj}	α_{max}
ITT(27J)			
= -3+14000B +800C +40Mo-40Ni -30Cu -11logCR₂-10$D_{15^\circ}^{-1/2}$-7RR	0.736	**0.721**	**0.002**
= -3+14000B +800C +40Mo-40Ni -30Cu -11logCR₂-10$D_{15^\circ}^{-1/2}$-7RR	0.736	**0.721**	**0.002**
= -20+13000B +800C-300Ti+45Mo-40Ni-35Nb-30Cu +6Mn+5Cr-11logCR₂-10$D_{15^\circ}^{-1/2}$-7RR+1logCR₁	**0.739**	0.715	0.858
0.5Kvmax			
= -22+15000B+1100C +35Mo-35Ni -30Cu+20Mn -15logCR₂-10$D_{15^\circ}^{-1/2}$	0.692	0.675	**0.010**
= -30+16000B+1100C-700Ti+35Mo-35Ni -30Cu+25Mn -15logCR₂-10$D_{15^\circ}^{-1/2}$-3RR-5logCR₁	0.702	**0.679**	0.206
= -30+16000B+1100C-800Ti+35Mo-35Ni+30Nb-30Cu+25Mn-2Cr-15logCR₂-10$D_{15^\circ}^{-1/2}$-3RR-5logCR₁	**0.702**	0.675	0.789



(a) Significances

(b) Effect in ºC

Figure 2. Plots of significances and effects of predictor variables on ductile-to-brittle impact transition temperatre (ITT), without considering $D_{15º}$.



(c) Significances

(d) Effect in ºC

Figure 3. Plots of significances and effects of predictor variables on ITT, considering $D_{15º}$.

From the first group (Table 4.a), it is remarkable the effect of *RR*, C and B, whose coefficient present the lowest significances ($\alpha << 0.1$), as shown in Figure 2.a. Concerning the significant effects (Autofit, $\alpha < 0.1$); C, B, Mo and Mn impair both *ITT* 27J and 0.5Kvmax. In opposite direction; **Nb**, Cu, Ni additions on the one hand, and increasing the level of *RR*, mainly, and CR_2 on the other, improve the toughness properties. Solely Ti shows no effect on both models, with significances well above 0.1. In the case of Cr and CR_1, they only have significative effect on *ITT* 27J and 0.5Kvmax, respectively. The Figure 2.b represents the effect (in ºC) of the proved predictor variables, where the columns represent the average effects and the error bars display the maximum and minimum observed effects.

The results from the second model (Table 4.b), where the $D_{15º}^{-1/2}$ has been included, are also shown in Figure 3. It is worthy noting the effect of grain size ($D_{15º}^{-1/2}$). This latter presents, by far, the strongest effect improving the toughness, as shown in Figure 4.

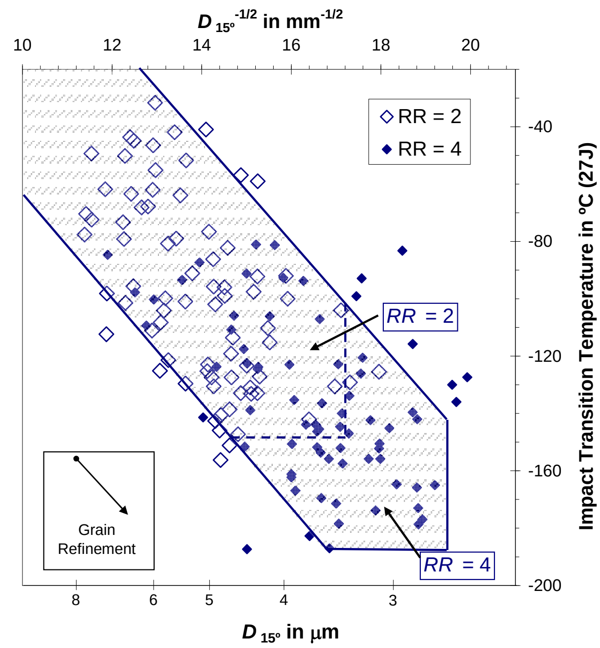
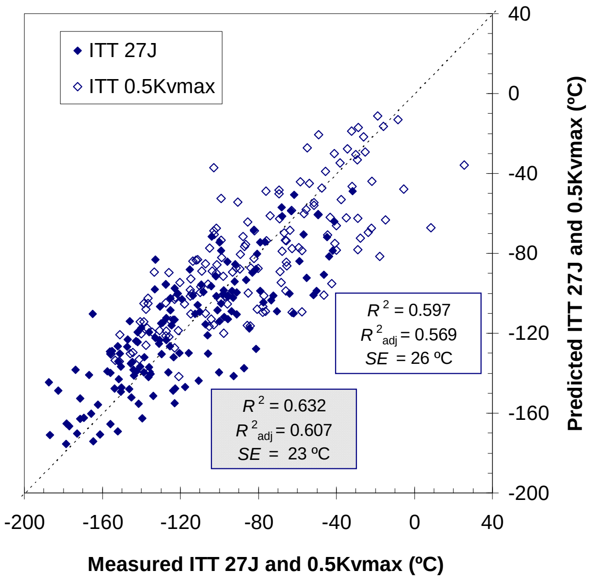


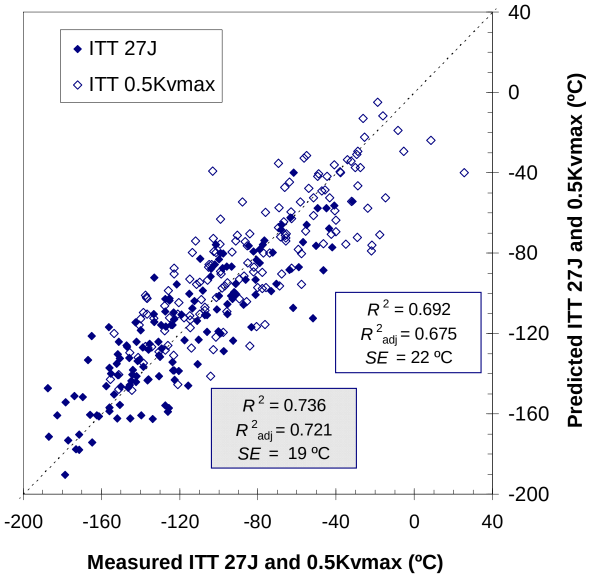
Figure 4. Effect of mean grain size of 15º ($D_{15º}$) and Rolling Reduction (RR) on Impact Transition Temperature (ITT 27J, ºC).

Plots of experimental versus predicted parameters were generated for the 26 casts used to compute the reponse equations considering only the Autofit method ($\alpha < 0.1$), as shown in Figure 5. Additional plots show the experimental versus the predicted values for 4 additional casts (industrial cast, Table 3) to validate the proposed equations, see Figure 6.

In the light of statistical parameters obtained from both model adequacy and model validation, the models with $D_{15º}^{-1/2}$ predict far more accurately. The determination coefficients (R^2) of toughness models improve substantially from about 0.63 and 0.60 to 0.74 and 0.70 for *ITT* 27J and 0.5Kvmax, respectively. Similar behaviour is found for adjusted coefficients (R^2_{adj}) since the response equations obtain more accurate predictions with a lower number of regressors, Figure 5.



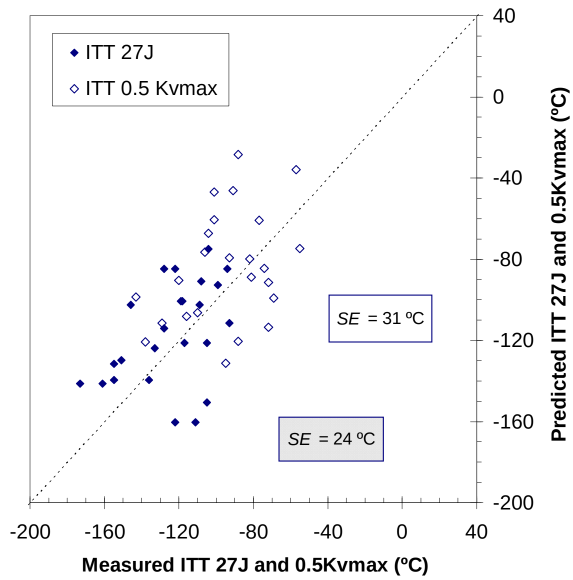
(a)



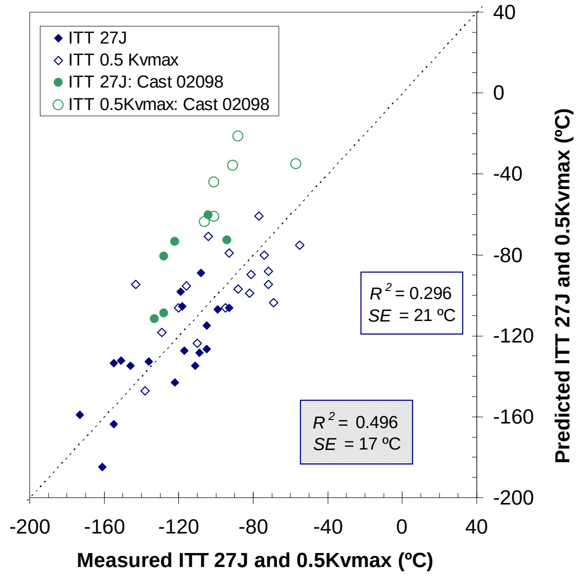
(b)

Figure 5. Experimental vs. predicted plots for toughness models (Model Adequacy), without considering $D_{15º}$ (a), and considering $D_{15º}$ (b). Autofit (26 casts).

Concerning model validation, the determination coefficients for the first models are negative, being, therefore, not included in the Figure 6.a. However, when considered the grain size ($D_{15º}$), the models predict reasonably well, with standard errors of 17 and 21ºC for *ITT* 27J and 0.5Kvmax, respectively.



(a)



(b)

Figure 6. Experimental vs. predicted plots for toughness model (Model Validation)s, without considering $D_{15º}$ (a), and considering $D_{15º}$ (b). Autofit (26 casts).

5.1. Comparison between response equations

Taking a glance to response equations (Table 4) and/or effect of predictor variables (Figure 2, Figure 3), when the $D_{15º}$ is considered or not, it is possible to draw the following conclusions, mainly for *ITT* 27J:

- ☑ In a previous publication, the effect of both the same rolling conditions and intelligent design of experiments on grain size was studied [7], giving place to the next response equation (Autofit):

$$D_{15^\circ} = 6 + 450B - 15C - 10Nb + 1Mn + 1Cr - 0.7RR - 0.2\log CR_2 \qquad (1)$$

☑ It is observed that the regressors coefficients which affect to D_{15^o}, such as B, C, Nb, Cr, CR_2 and *RR*, change substantially if this latter parameter is included or not, see Table 5, where such regressors are highlighted in grey. It is possible to say that the effect of these regressors can be partly included by the $D_{15^o}^{-1/2}$ term. Whilst on the other hand, the rest of regressors, such as Mo, Cu, and Ni which do not show proved effects on D_{15^o} hardly change.

Table 5. Comparison between toughness models.

ITT 27J - Autofit							
Without considering D_{15^o}				Considering D_{15^o}			
Term	Coefficient	Std Error	Significance	Coefficient	Std Error	Significance	Term
Ni	-37.2	9.7	2.0E-04	-35.8	8.2	2.2E-05	Ni
Nb	-182.6	95.8	0.059				Nb
Cu	-39.1	9.1	3.4E-05	-28.7	7.7	2.6E-04	Cu
$\log CR_2$	-6.5	2.6	0.013	-11.4	2.3	1.5E-06	$\log CR_2$
				-9.5	1.1	4.0E-15	$D_{15^o}^{-1/2}$
RR	-19.5	1.8	5.4E-20	-6.7	2.1	1.8E-03	*RR*
Constant	-126.5	17.9	6.4E-11	-3.1	14.2	0.826	Constant
$\log CR_1$							$\log CR_1$
Cr	19.1	9.5	0.045				Cr
Mn	23.1	8.0	0.005				Mn
Mo	53.1	13.9	1.9E-04	40.9	11.6	6.0E-04	Mo
Ti							Ti
C	562.0	110.2	1.1E-06	815.4	88.5	3.4E-16	C
B	21526.4	2912.5	1.1E-11	13573.8	2614.2	6.9E-07	B

☑ Concerning elements such as Nb, Cr and Mn on *ITT* 27J, its disappearance (null effect) when $D_{15^o}^{-1/2}$ is considered might indicate that their whole effects on toughness is somehow related directly by means of grain size. Nevertheless, these elements have a proved solid solution strengthening effect, because of which the toughness should have been affected. This apparent contradiction might be explained as follows: The D_{15^o} shows the more powerful effect on toughness, much more than other effects like precipitation strengthening or solid solution. Therefore, the effect of these elements, chiefly Nb and Mn, on solid solution may be negligible in comparison with their effect on grain size. As instance, it is well known the strong effect of manganese on strength but also on that of grain size (D_{15^o}) [7].The null effect on Mn on toughness is consistent with the work developed by F. B. Pickering: In the classical equation of impact transition temperature, there was no apparent effect of manganese because its effect was incorporated in the grain size [8].

☑ Similar conclusions can be drawn for 0.5Kvmax.

6. SUMMARY AND CONCLUSION

☑ On the basis of measured grain sizes (D_{15^o}) and Impact Transition Temperatures (*ITT* 27J and 0.5Kvmax), multiple regression models have been developed, alloying to quantify the effect of both rolling parameters and chemical compositions on toughness properties for thermo-mechanically rolled structural and pipe steels.

☑ The findings show how rolling reduction (*RR*) plays one of the most important roles on final grain sizes distributions, refining the final grain size (D_{15^o}). This refinement is translated in a remarkable drop of Impact Transition Temperatures (*ITT*).

☑ It was observed that the alloying elements with strong effects on hardenability, promoting low transformation temperatures products (bainite), impair the toughness (B, Mn and Mo). On the other hand, it is remarkable the effect of high niobium additions (up to 0.12%) improving the toughness by means of grain size refinement.

☑ The results have shown that excellent toughness properties can be obtained by using low carbon contents (< 0.8 %) and high niobium additions (up to 0.12 %) without the use of more expensive alloying elements like molybdenum and vanadium.

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REFERENCES

[1] F. B. Pickering, *Towards Improved Toughness and Ductility*, Climax Molybdenum, Co. Symp., Kyoto, 1971, p. 9.

[2] L. J. Cuddy, *Metall. Trans.*, 15ª, 1984, p. 87.

[3] M. H. Thomas and G. M. Michal, in H. I. Aaronson *et al.* (eds.), Solid-Solid Phase Transformation, *TMS-AIME*, Warrendale, P. A, 1981, pp. 469-473.

[4] J. Majta, R. Kuziak, M. Pretrzyk, *Journal of Materials Proccessing Technology*, Vol. 80-81 (1998), pp. 524-530.

[5] D. Bhattacharjee, J. F. Knott and C. L. Davis, *Metallurgical and Materials Transactions A*: Physical Metallurgy and Materials Science, vol. 35 A, nº 1, 2004, p. 121.

[6] K. Wallin, Modified Tanh fitting algorithm for Charpy impact data, ECOPRESS Research Seminar on economical and safe application of modern steels for pressure vessels, 19th and 20th of May 2003, Aachen, Germany.

[7] M. Pérez-Bahillo, B. López and A. Martín-Meizoso, "Efecto de la rutina de laminación en el tamaño de grano en aceros con alto contenido en niobio", *X Congreso Nacional de Materiales*, Donostia - San Sebastián, Vol. 1 (2008), pp. 343-346.

[8] F. B. Pickering: *Material Science and Technology*, Vol. 7: Constitution and Properties of Steels, Ed. by R. W. Cahn, P. Haasen and E. J. Kramer (1992), pp. 54.