Proceedings of the 5th International Symposium on Steel Science (ISSS 2017)
Nov. 13-16, 2017, Kyoto, Japan: The Iron and Steel Institute of Japan

The complementarity between combinatorial screening and fundamental understanding in exploratory research: a case study on lean maraging steels

Laura Moli Sanchez[1*], Nele Van Steenberge[1], Pedro Marques Spinola De Andrade[1], Lode Duprez[1]

[1] Arcelormittal Global R&D Gent, Pres. J.F. Kennedylaan 3, 9060 Zelzate, Belgium

Abstract: Compared to classical incremental product developments, breakthrough research with a long term objective might require a different approach as less reference data are available to start from. Indeed, when exploring new processes and unknown ranges of chemical compositions in order to achieve a leap in properties, available models loose their validity and trustworthiness, less literature or past experimental data exist, etc. In such a scenario, a screening approach in which large compositional ranges and multiple process conditions are tested in an accelerated manner is a viable option to quickly identify hot spots, *i.e.* combinations of chemical compositions and process conditions resulting in first attractive properties. In parallel, this screening exercise can still be accompanied by fundamental studies as the latter might give the orientation on how to continue the development efforts starting from the selected hot spots. In this contribution this approach is illustrated via a case study on lean maraging steels, where more than 1000 materials were tested in an accelerated manner and compositional hot spots were identified and selected via advanced data mining using neural networks. Simultanuously, fundamental knowledge was obtained on generic alloys with ternary compositions for which single type of intermetallic phases were expected. Here, merging and correlating the results of thermodynamic predictions, dilatometry and DSC experiments and advanced microstructural characterisation allowed to obtain a deeper insight in the formation kinetics and effects of each intermetallic phase seperately. This approach of combining fast initial data generation and first fundamental understanding allowed to launch next dedicated product developments in a much more reasoned manner and in a much shorter time frame.

1. COMBINATORIAL METALLURGICAL RESEARCH

Combinatorial research or high-throughput screening (HTS) is a method for scientific experimentation which was originally mainly used in drug discovery and proved its relevance to the fields of biology and chemistry. The basic principle is a high-throughput screening that allows a researcher to quickly conduct millions of chemical, genetic, or pharmacological tests, and as such to speed-up intelligent decision making in research. The approach is particularly relevant for research in domains where little prior knowledge is available, or where the parameter space is large and multi-dimensional.

In the last decade, combinatorial research has been more and more applied for metallurgical research. At first this was majorly done via thin film technologies for intrinsic system properties, but most recently as well via bulk methodologies for screening of structural properties. In both cases this is realised by the use of small scale techniques that allow fast synthesis and testing. As such, combinatorial metallurgy can accelerate the discovery of innovative alloys in exploratory research, e.g. by

- Easier finding of ‘hot spots’ in multicomponent systems via basic testing (Fig. 1). As an example, one can imagine a first wave of rough screening of 100 compositions of an alloy system to identify the interesting compositions (‘hot spots’). In a second wave, a refinement of compositions is tested around these hot spots, which can also evaluate different heat treatments. Only in a third wave, upscaling to larger laboratory experiments is done to evaluate performance on larger scale samples.
- Accelerated generation of heat maps visualising properties vs chemistry and process conditions (Fig. 1). The related database allows for enhanced predictive power when combined with e.g. neural networks.

* Corresponding author. E-mail: laura.molisanchez@arcelormittal.com, telephone: +32 9 345 12 71.

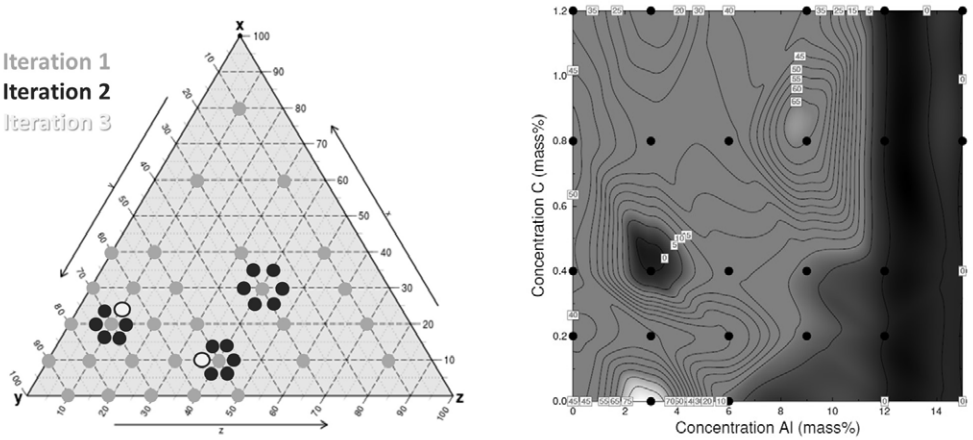

Figure 1: Two schematic illustrations of the power of combinatorial research: (left) detection and optimisation of 'hot spots' (right) accelerated property mapping as function of Al and C-content.

2. SCREENING LEAN MARAGING COMPOSITIONS

This paper discusses the case study of a research project that targeted a lean maraging steel with a strength level of 1400MPa and enhanced resistance to stress corrosion cracking (SSC). As SSC testing is quite expensive, a combinatorial approach was applied to come to a fast selection of chemistries and processings that could achieve the required strength level. In a first wave, around 1200 materials were synthesised, covering around 50 chemical compositions and multiple process conditions. For this purpose, small meltings were performed for each chemistry, followed by laboratory hot rolling in identical conditions. Subsequently, samples were re-austenitised, quenched and tempered in multiple T-t conditions. The chemistries were focussed on three families:

- A 'high Ni' family, based on the reference composition [1], for which Co and Mo reductions were evaluated
- A cost reducing 'low Ni' family.
- A Mn-family, starting from the work of Ponge et al [3].

For each chemistry, a complete mapping was done of the hardness evolution as function to tempering time and temperature. An example is shown in Fig. 2 for the reference 18Ni-0.5Ti-5Mo-9Co composition. The graph illustrates that the hardness target of ~43HRC could be achieved by a multitude of tempering conditions. Also, it shows clearly the accelerated hardening kinetics for higher temperating temperatures and the most stable tempering regime for 550°C.

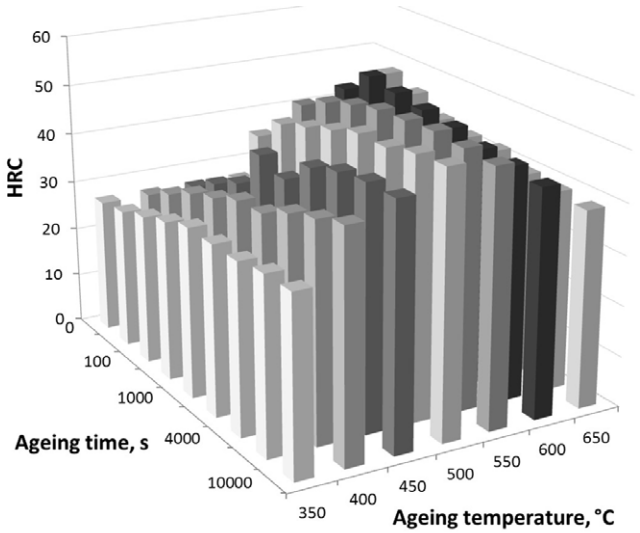

Figure 2: Hardness as function of tempering condition (time & temperature) for the reference maraging alloy18Ni-0.5Ti-5Mo-9Co. Similar mappings were generated for 50 different chemistries.

3. NEURAL NETWORK MODELLING

The power of combinatorial testing lies not only in the accelerated data generation, but also in the use of the large amount of data for predictive modelling. In this case of lean maraging steels, an artificial neural network was used to predict the hardness from chemistry and tempering conditions. The optimum neural network algorithm for the present study was the Levenberg–Marquardt algorithm. In this study, the artificial neural network model was implemented in Matlab®. It was found that the best fit was obtained for a model with a structure of 20 hidden nets and by using 70% of the data for the training, 15% for the validation and 15% for testing.

Fig. 3 shows some results of the neural network. Black points are the experimental data. The different grey shades represent the high Ni, low Ni and Mn families. Although the maximum hardness in the experiments did not exceed 69HRC, predicted data points with hardness higher than 69 HRC were found. Via this model, a second series of new chemistries was selected and tested in a second wave.

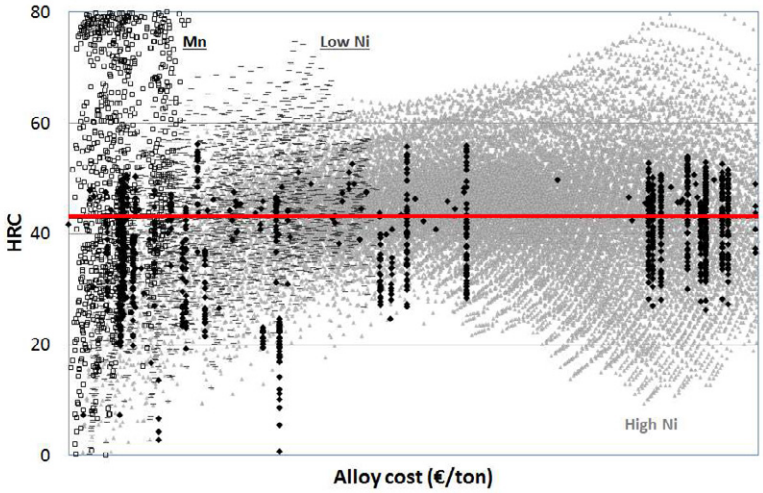

Figure 3: Predicted hardness based on the artificial neural network model as function of the alloy cost (aging conditions for the prediction: 550ºC-1h).

3. FIRST MICROSTRUCTURAL UNDERSTANDING

To understand the major tendencies that were observed in the experiments, high throughput XRD measurements and selected metallographic analysis were performed, Fig. 4. The optical images only allowed to get an idea of prior austenite grain size. Precipitation of intermetallics was only observed indirectly via a more severe etching attack of the (tempered) martensite. XRD graphs clearly allowed to monitor the formation of revert austenite for higher tempering temperatures.

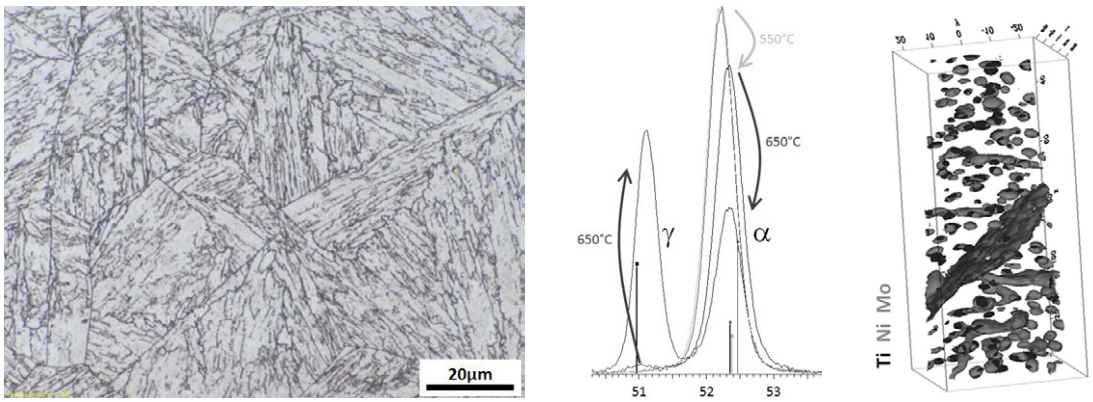

Figure 4: (left) LOM image in peak hardened condition at 550°C, (middle) XRD spectrum around the main α and γ peak, illustrating clearly the revert austenite transformation, starting already at 550°C, (right) APT data on the reference 18Ni-0.5Ti-5Mo-9Co alloy, showing the complex precipitation of multiple intermetallics.

4. PARALLEL FUNDAMENTAL INVESTIGATIONS

The large set of data generated by the combinatorial approach provides a good idea of the interesting compositions and processings and gives a first insight in microstructural evolutions. This does not necessarily exclude the need for a more fundamental understanding. As an example, the APT result in Fig. 4 illustrates the complexity of multiple intermetallic precipitation in the reference 18Ni-0.5Ti-5Mo-9Co alloy. To better understand the effect of each single element and intermetallic phase on microstructure and properties, generic ternary Fe-X-Y alloys were prepared to separate the various contributions.

Fig. 5 illustrates the results of a DSC measurement during the heating of a ternary Fe-18Ni-2,5Ti alloy, starting from the quenched martensitic microstructure in which it can be assumed that no intermetallics have precipitated during quenching. The overlay with Thermocalc predictions clearly shows a good correspondence, in particular for the revert austenite transformation. As such, a better understanding is obtained of the different phenomena that occur during heating, and the isolated effect on properties can be evaluated. Fig. 5 also shows the hardness evolution, which was obtained from interrupted trials in which samples were heated at the same heating rate as during DSC testing, followed by quenching after the characteristic temperatures of the DSC-curves. As such the strengthening contribution of each phase could be identified and intermetallic phases can be ranked on their contribution to properties. This work is still ongoing and extended to other properties than hardness.

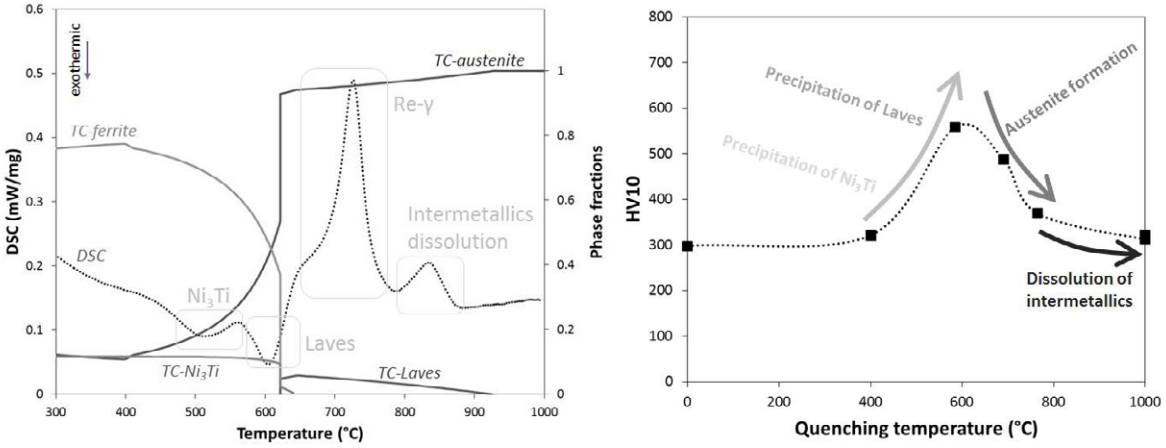

Figure 5: (left) DSC curve obtained during heating at 40K/min of a quenched Fe-18Ni-2,5Ti maraging superimposed on Thermocalc predictions (right) Understanding of hardness evolution during continuous heating.

5. CONCLUSIONS

In this contribution, a macroscopic view was given on combinatorial metallurgical research. The approach was used for a specific exploratory search towards a lean 1400MPa maraging steel with improved SSC resistance. Around 1200 materials were synthesised and characterised in 6 months time, which allowed to find first “hot spots”, i.e. interesting combinations of compositions and processings giving targeted strength levels. An artificial neural network allowed to define new, optimised compositions. Synthesis of generic alloys combined with advanced characterisation allowed to understand the formation kinetics of the respective intermetallic phases as well as the revert austenite transformation. All information allowed to launch a first upscaling on laboratory scale in which the most promising concepts are being tested more in detail on advanced properties.

REFERENCES

[1] A. S. M. Handbook, Volume 4: Heat Treating., ASM International, 72-74., 1991.
[2] W. Sun, “Development of Ultra-High Strength 'Lean' Maraging Steels,” Department of Materials Engineering, Monash University, Victoria, Australia, 2015.
[3] Raabe, D., Ponge, D., Dmitrieva, O., & Sander, B., “Nanoprecipitate-hardened 1.5 GPa steels with unexpected high ductility,” *Scripta Materialia,* vol. 60, no. 12, pp. 1141-1144, 2009.