# USE OF THE METHOD OF DESIGN OF EXPERIMENT FOR QUANTIFICATION OF THE INFLUENCE OF EXPERIMENTAL CONDITIONS ON THE CORRECTNESS OF THE MEASURED DATA BY DIRECT THERMAL ANALYSIS METHOD

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**Abstract**

Metallurgical processes of steel production are strongly connected with knowledge of the thermo-physical and thermodynamic properties of liquid metal during individual technological steps. Critical factors describing behaviour of steel in the process of its solidification and then important to a correct setting of conditions of steel casting and solidification of steel, which significantly affect the final quality of the as-cast steel, are liquidus ($T_L$) and solidus ($T_S$) temperatures.

Experimental determination of these temperatures (or generally additional phase transformation temperatures) can lead to the refinement of all these phase transformation temperatures as well as to a more precise determination of the interval of steels' solidification. Utilization of this approach, e.g. in the process of optimizing the technology of ingot or continuous casting, could lead to achieve more accurate results moreover in direct connection with numerical simulations, and thus a better knowledge of the impact of optimized technological parameters on the resulting quality of cast steel.

The paper is devoted to the application of method of Design of Experiment (DOE) to analyse and clear quantification of the impact of experimental conditions on the experimental results (measured data) of direct thermal analysis method (DirTA). First of all, it is a way to find a new approach to quantify and verify a statistical significance of experimental effects (influence of experimental conditions) on obtained phase transformation temperatures. The developed method allows filtering relevant data from individual thermo-analytical experiments for final calculation of high temperature phase temperatures more precisely than it was possible ever before. This is crucial for proper liquidus and solidus and liquidus temperatures' recommendation to real conditions.

**Keywords:** Steel, nickel, direct thermal analysis, design of experiment, experimental conditions

## 1. INTRODUCTION

Method DOE (Design of Experiments) means a gradual, conscious setting of selected controllable input factors and monitoring the impact of these factors on the output parameters (response) [1]. The combination of input factors is an optimized mathematical model in which maximum resolution can be achieved for the significance of factors and their interaction with as small a step as possible and a test experiment. DOE goal is to design and implement a plan of the experiment, by which it can be determined whether a particular factor or combination of these factors influences the monitored variables, or to find a level of factors which allow reaching / finding the optimum value of the monitored quantity. DOE allows study the effects of several factors by testing at various levels [2].

DOE in this case to the area of experimental studies of phase transformation temperatures of the steel during its solidification is applied. Specifically, it is used to refine the liquidus and solidus temperature in real industrially produced steel grades (continuously cast billets / ingots), which were determined by high-temperature Direct Thermal Analysis method [3]. It is obvious that the use of knowledge of actual liquidus temperatures can significantly influence the process of optimizing the production of ingots as well as continuously cast billets, both in the direct treatment of casting temperatures or in the setting of boundary conditions of numerical simulations [4,5]. Verification of the usability and implementability of the results in industrial practice can contribute to optimize the casting and solidification of steel.

## 2. SPECIFIC RESEARCH OVERVIEW, SPECIFICATION OF RESEARCH FOCUS

For this purpose, up to 26 steel grades (ingots and continuous cast billets) by the DirTA method were measured and the temperatures of liquidus and solidus (on experimental background of analyses in the **Figure 1**) were determined. An overview of the analysed steel grades in accordance to European standards and API standards is shown in **Table 1**.

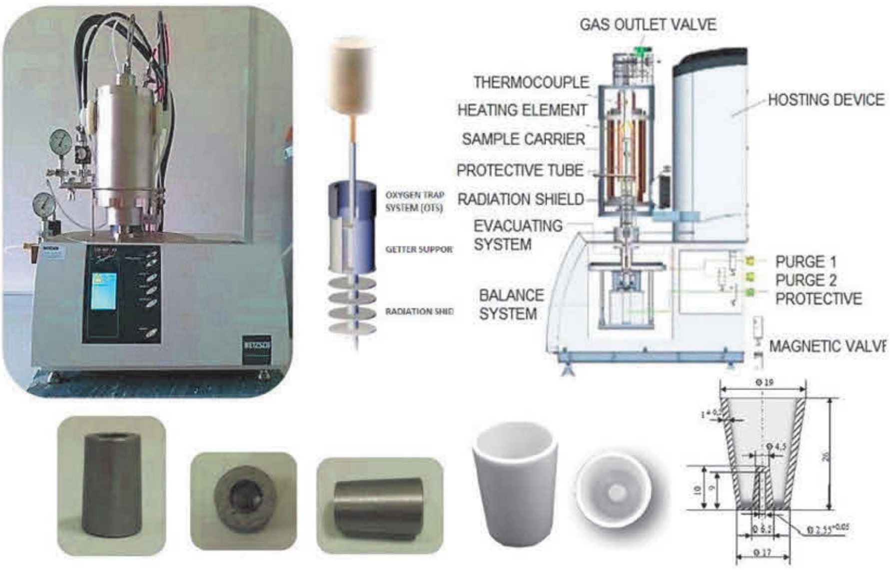


**Figure 1** Experimental background of analyses

**Table 1** Overview of the analysed steel grades

| Continuously cast billets | Ingots |
|---|---|
| ▪ steel samples from continuous cast billet with small and large diameter (Ø130 - 400 mm) | ▪ samples from steel ingots used in conditions of processing of forgings |
| ▪ steel for tubular steel production in oil fuel and natural gas extraction industry (oil fuel / gas pipelines, casing pipes); steel used in conditions of power industry (boiler tubes) | ▪ thick steel plates, blocks, pads, bars designed for special machine engineering elements from tool steel, bearing steel |
| ▪ steel designation according to European standards: EN 10280-1, EN10210-1, EN 10216-1. EN10216-2, EN10224, EN10297-1 | ▪ steel designation according to European standards EN 10027-1: designation according to chemical composition of |
| ▪ steel designation according to API standards: API 5L - steel grade A, B, X42, X46, X52, X56, T95; API 5 CT - steel grade J55, K55, L80-1, N80-1, N80Q, P110, Q125; SAE 1526; SAE 1518; SAE 1527; SAE 4125 | ▪ samples of steel grades 100CrMo6, 100CrMo7, X37CrMoV5-1, 56NiCrMoV7, 40CrMnNiMo8-6-4, X40CrMoV5-1, X37CrMoV5-1, 51CrV4, 34CrAlNi7, 40CrMnMo7 |

## 3. METHODOLOGICAL EXPERIMENT OF PURE NICKEL STANDARD

In relation to refine the liquidus and solidus temperatures of real steel grades, the series of methodological experiments on pure standard metal (nickel) were preceded [3]. The purpose of those methodological experiments was principally a verification of the suitability of the use the DirTA method and used experimental arrangement (experimental system) to generating relevant and reproducible results, and then to quantify the value the temperature correction for experiments of real steel samples. But, with thus also relates the further study of the behaviour of the pure standard metal during the experiment and to identify the effects of the experimental conditions on the experimental phase transformation temperatures.

Now, in addition to the standard approach, the effects of experimental conditions on the value of the melting point or solidification point temperatures of pure nickel by application of the method of Design of experiments (DOE) were quantified. After the identification the statistical significance of studied influential factors (some key effects) of experimental conditions and detection of the deviations on the pure standard, DirTA method, including the DOE application of the entire series of experiments on real steel grades was applied.

### 3.1. Use DOE on a pure nickel standard

Method of planned experiment DOE to the evaluation, resp. more precisely quantification of the statistical significance of effects of these selected factors of experimental conditions were used:

- statistical significance of sample mass influence,
- statistical significance of heating or cooling rate influence,
- statistical significance of the experimental mode influence (linear heating / linear cooling conditions),
- statistical significance of the influence of the using the sequences in DirTA experiments.

Design of Experiment (DOE) in S*TATGRAPHIC Plus* software was realized.

### 3.2. Plan of DOE: Statistical significance of the effect of sample mass and heating rate to melting point and solidification point temperature

In accordance with the theory of DOE [1,2], a melting point or solidification point temperature of the nickel standard were successively chosen as the study response (the quality indicator (Y)). Factors influencing the defined response are: the weight of the sample (M) and the heating rate ($R_{heat}$), and follow the cooling rate ($R_{cool}$). In this case of DOE construction focused on evaluation the statistical significance of the effect of sample mass and heating rate, the two DOE application were provided. The first one was - planned experiment of the nickel samples of weights 10 and 20 g (**Table 2)** and the second one was DOE construction at a reduced sample mass interval (17 and 20 g), **Table 3**.

**Table 2** Overview of the factors and levels used for DOE construction

| Factor | Mark of factor | Lower level | Upper level |
|---|---|---|---|
| sample mass (g) | M | 10 | 20 |
| heating rate, resp. cooling rate (°C·min$^{-1}$) | $R_{heat}$, resp. $R_{cool}$ | 5 | 20 |

For first DOE application - an evaluation of the statistical significance of the sample mass and consequently the effect of heating or cooling rate, represent the lower and upper levels of the observed factors:

- sample mass (M) - two extreme weights of nickel sample (lower and higher, it means 10 g and 20 g) for which DOE was assembled,
- in case of heating and cooling rate effects, the lower and upper factor levels correspond to two different heating or cooling rates. Again one lower (5 °C·min$^{-1}$) and one higher (20 °C·min$^{-1}$).

And then, one more application of DOE (the second one) for the same combination of factors nonetheless at a reduced sample mass interval (17 g and 20 g) with the using already identical heating or cooling rates as in the first case of realized DOE (with samples weighing 10 g and 20 g), were designed, **Table 3**.

**Table 3** Overview of the factors and levels used for DOE construction at a reduced sample mass interval

| Factor | Mark of factor | Lower level | Upper level |
|---|---|---|---|
| sample mass (g) | M | 17 | 20 |
| heating rate, resp. cooling rate (°C.min$^{-1}$) | $R_{heat}$, resp. $R_{cool}$ | 5 | 20 |

Correspondingly to the previous DOE construction, the lower and upper factor values represent:

- sample mass (M) - new selected mass of nickel sample (one lower and higher: 17 g and 20 g),
- the heating and cooling rate ($R_{heat}$, $R_{cool}$) included in the planned experiment, again one lower one (5 °C·min$^{-1}$) as lower level and one higher (20 °C·min$^{-1}$), which was the upper level of the DOE.

With regard to a sufficient number of experiments for all combinations of M and $R_{heat}$ (or $R_{cool}$) levels, complete two-level full factorial designs using the measurable (numerical) description of the variables (factors) were prepared for each response (**Table 4 - Table 7**).

Individual DOEs differ in Y-values, determined by DirTA, esp. in linear heating conditions, resp. linear cooling (melting point temperature, resp. solidification point temperature) and in sample mass interval.

**Table 4** DOE in STATGRAPHIC Plus software: statistical significance of effect of sample mass and heating rate to melting point temperature

| Run | M | $R_{heat}$ | Y (melting point) |
|---|---|---|---|
| $YY_1$ | 10 | 5 | 1,449.0 |
| | 20 | 5 | 1,449.0 |
| | 10 | 20 | 1,452.0 |
| | 20 | 20 | 1,450.4 |
| $Y_2$ | 10 | 5 | 1,451.0 |
| | 20 | 5 | 1,449.0 |
| | 10 | 20 | 1,450.3 |
| | 20 | 20 | 1,451.0 |

**Table 5** DOE in STATGRAPHIC Plus software: statistical significance of effect of sample mass and cooling rate to solidification point temperature

| Run | M | $R_{cool}$ | Y (solidification point) |
|---|---|---|---|
| $Y_1$ | 10 | 5 | 1,444.1 |
| | 20 | 5 | 1,448.4 |
| | 10 | 20 | 1,445.5 |
| | 20 | 20 | 1,447.4 |
| $Y_2$ | 10 | 5 | 1,439.7 |
| | 20 | 5 | 1,445.5 |
| | 10 | 20 | 1,441.4 |
| | 20 | 20 | 1,446.3 |

**Table 6** DOE in STATGRAPHIC Plus software: statistical significance of effect of sample mass at a reduced sample mass interval and heating rate to melting point temperature

| Run | M | $R_{heat}$ | Y (melting point) |
|---|---|---|---|
| $YY_1$ | 17 | 5 | 1,448.7 |
| | 20 | 5 | 1,449.0 |
| | 17 | 20 | 1,448.2 |
| | 20 | 20 | 1,450.4 |
| $Y_2$ | 17 | 5 | 1,445.0 |
| | 20 | 5 | 1,449.0 |
| | 17 | 20 | 1,450.9 |
| | 20 | 20 | 1,451.0 |

**Table 7** DOE in STATGRAPHIC Plus software: statistical significance of effect of sample mass at a reduced sample mass interval and cooling rate to solidification point temperature

| Run | M | $R_{cool}$ | Y (solidification point) |
|---|---|---|---|
| $Y_1$ | 17 | 5 | 1,445.2 |
| | 20 | 5 | 1,448.4 |
| | 17 | 20 | 1,445.2 |
| | 20 | 20 | 1,447.4 |
| $Y_2$ | 17 | 5 | 1,441.8 |
| | 20 | 5 | 1,445.5 |
| | 17 | 20 | 1,444.6 |
| | 20 | 20 | 1,446.3 |

### 3.3. Plan of DOE: statistical significance of the effect of the temperature of experimental mode and using the sequences in the DirTA experiments on the melting point and solidification point temperature

In accordance with the theory of DOE [1,2], the planned experiment of DOE for new selected influential factors were designed. As defined responses (Y), either the melting point or the solidification point temperatures was chosen. Statistical significance of effect of temperature mode (R) in which the phase transformation temperature was determined (under the linear heating conditions (H), or under the linear cooling conditions (C)), and from which sequence (S) of the experiment (from 1st or 2nd cycle). The factors influencing the defined quality indicator (Y) and whose significance were detected by DOE, is shown in **Table 8**.

**Table 8** Overview of the factors and levels used for DOE construction

| Factor | Mark of factor | Lower level | Upper level |
|---|---|---|---|
| regime of experimental mode | R | linear heating (H) | linear cooling (C) |
| turn of sequence | S | 1st cycle | 2nd cycle |

In this case of DOE construction, the lower and upper factor values represent:

- experimental mode is represent by the type of temperature mode: the lower level of the factor indicating the temperature type of experimental mode represents linear heating mode (H) and the upper level represents linear cooling mode (C),
- the turn of sequence is represent by 1st cycle of experiment run as lower level and by 2nd cycle of experiment run as upper level of studied factor.

Based on definition of the factors and individual lower and upper levels using the categorical description of the variables (factors), a full factorial design of DOE on two-levels for all combinations of R and S to examined response was design (**Table 9**).

**Table 9** DOE in STATGRAPHIC Plus software: statistical significance of effect of the regime of temperature mode sample mass and turn of sequence to melting point or solidification point temperatures

| Run | R | S | Y (melting point or solidification point) |
|---|---|---|---|
| $YY_1$ | H | 1 | 1,444.0 |
| | C | 1 | 1,445.7 |
| | H | 2 | 1,446.0 |
| | C | 2 | 1,440.2 |
| $Y_2$ | H | 1 | 1,445.0 |
| | C | 1 | 1,444.4 |
| | H | 2 | 1,446.2 |
| | C | 2 | 1,442.1 |

### 3.4. DOE results and discussion

Statistical significance of all above defined and explained factors is tested at a significance level $\alpha$ = 0.05 for a confidence level of 95 % through a P-value that indicates in STATGRAPHIC Plus software terminology their statistical significance. The critical significance value is 0.05. If the P-value effect is less than the critical significance value (i.e. 0.05), the effect of the observed factor is statistically significant. As the model captures the individual data, the R-squared value (adjusted to degree of freedom) tells us how the model computes the data.

**Estimated effects for nickel standard due to evaluation the statistical significance of the effect of sample mass and heating or cooling rate to melting point and solidification point temperature**

Chosen factors (sample mass and heating and cooling rate) were tested for statistical significance for each of their effects, **Table 10**.

From discussion focused on study of mass and heating rate effect on melting point temperature for wider mass interval <10;20> grams (**Table 10**) it can be said, that for combination of studied factors wasn't indicated statistical significant effect. This conclusion was confirmed by P-values of these effects, which are higher than critical significance value (0.05). However, a combination of factors of sample mass and cooling rate to solidification point and DOE result proved the statistical significance of sample mass analysed under the linear cooling conditions (P-value 0.011) and this DOE model fits the data file very well (90.35 %).

If focused on study of mass and heating/cooling rate effect on melting/solidification point temperature for reduced mass interval <17;20> grams (**Table 10**), it can be stated that results are similar to above discussed wider mass interval <10;20>. Only during cooling regime exists statistical influence also of sample mass on final solidification point temperature. R-squared values are also close to above discussed.

**Table 10** Analysis of statistical significance of effects of studied factors

| Mass interval (g) | Studied factors | P-value | | Statistical significance | R-squared adjusted value |
|---|---|---|---|---|---|
| <10;20> | sample mass and heating rate | sample mass | 0.4139 | NO | 30.65 % |
| | | heating rate | 0.1599 | NO | |
| | | interaction | 0.7435 | NO | |
| | **sample mass and cooling rate** | **sample mass** | **0.011** | **YES** | **90.35 %** |
| | | cooling rate | 0.4043 | NO | |
| | | interaction | 0.351 | NO | |
| <17;20> | sample mass and heating rate | sample mass | 0.3036 | NO | 29.12 % |
| | | heating rate | 0.1972 | NO | |
| | | interaction | 0.7324 | NO | |
| | **sample mass and cooling rate** | **sample mass** | **0.0285** | **YES** | **82.26 %** |
| | | cooling rate | 0.4093 | NO | |
| | | interaction | 0.3503 | NO | |

**Estimated effects for nickel standard: evaluation the statistical significance of the temperature regime of experimental mode and use the sequences in the DirTA experiments on the melting point and solidification point temperature**

In the case evaluation the statistical significance of the temperature regime of experimental mode and use the sequences in the DirTA experiments on the melting point and solidification point temperature, 2 effects have a P value of less than 0.05, meaning that they are significantly different from zero at the 95 % confidence level (**Table 11**).

**Table 11** Analysis of statistical significance of effects of studied factors

| Studied factors | P-value | | Statistical significance | R-squared adjusted value |
|---|---|---|---|---|
| regime of experimental mode and turn of sequence | **linear heating or linear cooling** | **0.0478** | **YES** | **84.20 %** |
| | cycle | 0.1888 | NO | |
| | **interaction** | **0.0271** | **YES** | |

Among statistically significant effects belong the regime of linear heating or linear cooling mode with P-value=0.0478 and interaction of studied factors with P-value=0.0271. The R-squared statistic (R-squared adjusted value=84.20 %), which is comparing models with different numbers of independent variables, indicates that the model as fitted explains 84.20 % of the variability in melting or solidification point temperature.

## CONCLUSION

Method of planned experiment DOE more precisely quantified the statistical significance of the key effects of studied factors of experimental conditions. Previous methodological experiments of DirTA method on pure nickel standard carried out some general conclusions, which standardized the setup of DirTA experiments. These theoretical assumptions [8] on pure nickel standard, the evaluation of statistical significance of sample mass influence and heating cooling rate influence by DOE application were confirmed:

- A combination of factors: sample mass and cooling rate proved in consistence with the statistical significance of sample mass analysed under the linear cooling conditions on solidification point

(**Table 10**); it is therefore appropriate to maintain stable mass of the studied samples (20 grams and higher) to eliminate the sample mass effect,

- Based on previous knowledge and to be close to equilibrium conditions the lowest heating/cooling rate was selected for real steel samples $T_L$ and $T_S$ study, although no statistically significant influence of the rate of heating was identified.

Further DOE application corresponding with physically-chemical knowledge of the background of DirTA experiments to comparing temperature mode and cycle, reps. statistical significance of the experimental temperature mode influence (linear heating and linear cooling conditions) and statistical significance of the influence of using the sequences in DirTA experiments carried out the most interesting findings in relation with DirTA analyses on real steel samples. The impact of effects of temperature mode applied on pure nickel standard in combination with different cycles show how such conditions works:

- DOE results (**Table 11**) on nickel standard have detected the statistical significance of temperature mode. This conclusion is important in context with quantification the value of temperature correction to real materials (real steels) should be performed temperature correction separately for heating and cooling conditions.
- The sequences are not statistical significant individually, but only in interaction with temperature mode (**Table 11**), that mean probably no apparent any significant changes of material properties of pure nickel between first and second cycle of experiment, which would be demonstrate a significant impact on the experimentally determined melting or solidification point temperature.

This approach would be therefore appropriate to apply to real materials (real steel samples) and conclusively for each steel grade provided in accordance with the principles of the planned experiment to give this significance identified, because we cannot automatically supposed, that it will behave the heterogeneous material as steel just as pure nickel standard. That is also related to define the access to averaging and pooling achieved liquidus and solidus temperatures and determination whose values would be used as a mean value of liquidus and solidus temperature, which mean finally quantification of one proper liquidus and solidus temperature. Final quantification of one proper liquidus and solidus temperature represents our further task. Application of DOE allows the most simply way for identifying the statistical significance of the key effects of experimental conditions and their more precisely refinement moreover without time and additional financially demanding and large material analysis and without the use the large data file and work with data, which have technological context corresponding with studied purpose and methodological background.

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## REFERENCES

[1] KUEHL, R.O. *Design of Experiments: Statistical Principles of Research Design and Analysis*. 2nd ed. Belmont, California: Brooks/Cole, 2000. p. 666.

[2] MONTGOMERY, D.C. *Design and Analysis of Experiments*. 5th ed. USA: John Wiley & Sons, Ltd., 2001. p. 684.

[3] GALLAGHER, P.K. *Handbook of Thermal Analysis and Calorimetry: Principles and Practice.* Amsterdam: Elsevier Science B.V., 1998. p.691.

[4] ZHANG, Q., WANG, X. Numerical simulation of influence of casting speed variation on surface fluctuation of molten steel in mold. (In:) *Journal of Iron and Steel Research, International*. 2010. vol. 17, no. 8, pp. 15-19.

[5] GRYC, K., STROUHALOVÁ, M., SMETANA, B., SOCHA, L. and MICHALEK, K. Influence of direct thermal analysis experimental conditions on determination of the high temperature phase transformation temperatures. *Archives of Metallurgy and Materials*. 2015. vol. 60, no. 4, pp. 2867-2871.