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Weld-metal Property Optimization from Flux Ingredients Through Mixture Experiments and Mathematical Programming Approach

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This paper presents a new methodology for weld-metal properties optimization from welding flux ingredients. The methodology integrates statistical design of mixture experiment with mathematical programming optimization technique. The mixture experiment is responsible for the modeling of the weld-metal properties as a function of welding flux levels while mathematical programming optimizes the model. Data and confirmed models from the literature were used to perform optimization on the responses. The maximum values possible with the prevailing conditions for acicular ferrite, charpy impact toughness and silicon transfer are 51.2%, 29 J and 0.231% respectively while the minimum oxygen content possible is 249 ppm. The new methodology is able to eliminate the limitations associated with the traditional experimental optimization methodology for flux formulation.

Keywords: *welding flux, extreme vertices design, mathematical programming, optimization*

1. Introduction

Advanced materials are being developed to improve energy efficiency, corrosion resistance, high temperature performance, cryogenic performance and mechanical properties of the many industries of the future. Most of these materials are designed to be weldable; hence their effective deployment is highly dependent upon the development of welding technology. Quintana et al.[1] observed that rapid deployment of these materials are hampered because arc welding technology has not been able to keep pace with the development of these new materials. Welding flux design is one of the key areas of arc welding technology that require improvement because the weld-metal quality, productivity of the welding process and economical weld production depend largely on the flux formulation. Operational characteristics such as arc initiation and stability, minimum spatter, positional welding, high deposition rate, penetration and bead morphology are influenced by the welding flux formulation[2-8]. The quality of weld-metal is often evaluated by many characteristics such as chemical composition, mechanical properties, bead profile and microstructure. Studies have shown that these characteristics are influenced by the welding flux formulation; therefore it is important to select the right type of welding flux ingredients and choose the appropriate proportions of the various flux ingredients to attain a good weld-metal quality[2-10].

The conventional approach to welding flux development is by experimental optimization. Although experimental optimization is based on experience, the principles of physics, chemistry, and metallurgy, it is often difficult to know a priori how the flux ingredients interact to determine the operational characteristics of the flux and the final properties of the weld-metal. It is also difficult to know a priori or early at the experimental stage whether it is feasible to achieve the desired performance level with the flux ingredients being used or not until after much resources and considerable efforts have been expended on experiments. Such situations results in expensive and extensive trial and error experiments because the flux formulator has to drop some ingredients and introduce new ones and then continue with the trial and error experiments until a suitable flux formulation is achieved. Experimental optimization approach is therefore very slow and costly and as a result lags behind the development of new materials. The consequence is that rapid deployment of new materials is hampered[1,11]. In addition, the welding flux developed from such methods has a random character and often far from optimal because the best among the experimental flux formulations is usually selected as the optimum flux for the given metal under the given welding conditions[2,12]. Since it is not practical to explore all combinations of flux compositional variations due to time and cost limitations, the best experimental flux formulation can not be guaranteed to be the optimum. The ability to develop welding flux with lesser number of experiments and identification of the optimal flux formulation for the needed performance level is an enabling technology that will address the need for rapid deployment of new materials.

Recent research efforts are directed towards reducing the number of experiments and as a consequence reduce the lead time and time to market a new welding flux and at the same time achieve an optimum formulation. Ren et al.[13] tried to overcome these problems with a new approach to experimental optimization. They used a design of experiment method (DoE) known as uniform design (UD) to develop a new agglomerated flux for high speed and multi-arc SAW. In the UD, the only thing to be considered is the uniform dispersion of the experimental points in the experimental space. The reason for their use of the UD was that since the experimental points are uniformly scattered, they are more representative of the whole experimental region and as a result the optimum results in the designed experiments will not be far from the global optimum for the whole experimental space.

The UD approach reduces the amount of experimental efforts. However, the optimum result from the UD experiment can not be guaranteed to be the global optimum within the experimental space. Although the UD method has advanced the traditional experimental optimization, the result may be suboptimal or at best near optimal. Even if par chance the optimum result from the UD experiment coincides with the optimum in the total experimental space, there is no quantitative means (optimality criteria) for its identification. The

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means by which the real optimum flux composition may be identified is to develop empirical models from the results and integrate it with optimization techniques appropriate for their solution.

Kanjilal et al.[5-8] used another form of DoE technique known as the extreme vertices design (XVERTD) proposed by McLean and Anderson[14]. In the XVERTD technique, the constraints on the q flux ingredients define the experimental region, which is usually a (q – 1) dimensional simplex. The extreme vertices of the simplex, the centroids of each of the faces and the centroid of the entire simplex are determined and used as the experimental points or treatment combinations. The XVERTD method is a proven approach that is sufficient not only to fit the proposed model but also allows a test of model adequacy from a minimal experimental efforts[14]. Kanjilal and his co-investigators[5-8] used their experimental data to develop prediction models for the measured responses such as weld-metal composition, mechanical properties, microstructure and element transfer characteristics of the flux. However, they did not use the models to perform optimization on the responses. To be able to perform optimization on the responses, the XVERTD methodology should be coupled with mathematical programming optimization techniques. Usually, the aim of optimization in welding flux design is to minimize, maximize or hit the target of a response(s). With such models the formulation of welding flux can now be based on quantitative footing and advance the state-of-the-art of welding flux technology. With the integration of MP optimization techniques with XVERTD, the limitations of experimental optimization can be addressed, particularly, the random character of welding flux can be eliminated and optimality guaranteed. Also, it will be possible to ascertain the feasibility or otherwise of achieving the required operational characteristics and weld-metal performance level with the flux ingredients early before much effort is expended on experiments. Reduction in costs of labour, materials and energy associated with extensive experimental weld production and testing coupled with reduction in lead-time for the new flux are also benefits of such MP optimization models.

Although MP optimization techniques are not new in arc welding technology, to the best of our knowledge, its application to welding flux formulation has not appeared in the open literature. In this paper, we demonstrate how MP can be integrated with the XVERTD for the determination of flux ingredient levels that optimizes the desired responses of the Flux Designer (FD) using data from the literature. Because the integration of MP with XVERTD for welding flux formulation is novel, we give a brief systematic procedure for its application. Next, a numerical example is solved using the confirmed models of Kanjilal and his coinvestigators[5-8].

2. Description of the Combined XVERTD and MP Methodology

Published literature suggests that factorial design is not suitable for welding flux formulation because welding flux is a mixture of ingredients and the final properties of the flux depend on the relative proportions of the ingredients in the mixture[11]. Experiments where the input factors are the ingredients/components of a mixture and the properties (response variables) are functions of the proportions of the ingredients are known as mixture experiment. XVERTD as a type of mixture experiment is a DoE method that allows the experimenter to establish the mathematical relationship between input factors (mixture components) and the response variables with minimal number of experiments. The details of the XVERTD method are beyond the scope of this paper. The details are presented in the articles of McLean and Anderson[14], Snee and Marquardt[15] and Ding et al.[16]. The combined XVERTD and MP method are in 2 phases; the first phase deals with model development while the second phase involves using the developed model to perform optimization.

MP optimization techniques have been available for some time but their application in welding flux design is sparse. Because the application of MP in welding flux development is new, we present a brief description of its procedure before the discussion on the specific steps to be taken in coupling it with the XVERTD. The development of MP models generally involves the following: 1) Identification of the variables of the model. In welding flux design the most commonly encountered variables are the levels/proportions of the flux ingredients. 2) Identification of the structural/technological constraints of the problem and the development of their mathematical expressions. For the case of welding flux, the lower and upper bounds on the levels of flux ingredients are the constraints. Other constraints may be added depending on the situation. 3) Identification of the quality characteristics (response variables) and the preferences of the FD. For instance, he may wish to maximize a desirable response or minimize an undesirable response. 4) Construction of the response function in terms of the variables (proportions of flux ingredients). 5) Solving the model using appropriate computer software.

2.1. Phase I

The FD plans the experiment, conducts the experiment and uses the data from the experiment to fit a regression model according to the XVERTD methodology. The specific steps are[11,17,18]:

a) Definition of the objectives of the experiment;
b) Identification of important welding flux ingredients (input variables);
c) Identification of response variables to be measured (mechanical properties, microstructure, chemical composition, etc…);
d) Finding the upper and lower limits of the various flux ingredients. This is usually through some preliminary experiments or data from published literature;
e) Development of the design matrix by the McLean and Anderson algorithm or by any other appropriate algorithm. The McLean and Anderson[14] algorithm selects the extreme vertices of the experimental region, the centroids of the faces and the overall centroid of the simplex defined by the constraints as the treatment combination sufficient to fit model that relates the input variables to the response variables;
f) Conducting the experiment as per the design matrix;
g) Measuring and recording the responses;
h) Proposing an appropriate model for the relationship between the flux ingredients and the response variables;

$$\eta_k = f_k(x_1, x_2, ...x_q), k = 1, 2, ...k \quad (1)$$

where η_k is the k[th] response variable, q is the total number of flux ingredients, K is the total number of responses and x_1, x_2, ...x_q are the input variables (i.e. proportions of flux ingredients). Some of the common analytical model forms are presented by Adeyeye and Oyawale[11];

i) Calculation of the coefficients of the polynomials;
j) Checking the adequacy of the model developed;
k) Arriving at the final mathematical models;
l) Conducting the confirmatory test; and
m) Using the model to predict the value of response variable(s) from a given combination of flux ingredients.

2.2.Phase II

At this phase, the FD uses the confirmed models of phase I to perform optimization of the response(s)

The steps are;

a) Selection of the response variable(s) to be optimized from the identified responses of phase I;
b) Using the confirmed model(s) of the selected response variables of step (i) as the objective functions to be optimized.
c) Construction of the constraints of the model. The most frequently encountered constraints in flux formulation are given in equations (2) and (3) below

$$0 \le L_i \le x_i \le U_i \le 1,\ i = 1,2,...,q \quad (2)$$

$$\sum_{i=1}^{q} x_i = 1 \quad (3)$$

where
x_i: The proportion of the i[th] ingredient in the flux
L_i: The lower bound of the i[th] ingredient
U_i: The upper bound of the i[th] ingredient
q: The total number of ingredients.
The first constraint keeps each mixture component proportion between the lower and upper bounds and the second constraint makes sure that at any point in the mixture space, the total sum of the proportions of all the components adds up to unity. Depending on the situation other forms of linear multi-component constraints may be added.
d) Solving the model to determine the relative proportions of flux ingredients that optimizes the desired response(s) i.e.Maximize or Minimize, $\eta_k = f_k(x_1, x_2, ..., x_q)$ Subject to:

$$0 \le L_i \le x_i \le U_i \le 1,$$

(4); and

$$\sum_{i=1}^{q} x_i = 1$$

e) Using the results to formulate the welding flux that meets the desires of the FD.

3. Numerical Examples

Kanjilal et al.[5-8] have used the XVERTD to develop empirical models for the prediction of weld-metal properties as a function of welding flux ingredients for the submerged arc welding of C-Mn steel. The design matrix according to the XVERTD and results of their experiments are shown in Table 1 while the empirical models are shown in Table 2. The flux ingredients used were the reagent grade CaO, MgO, Ca_2F and Al_2O_3. The experiments were conducted at fixed welding parameters. In this study we couple MP optimization technique with XVERTD by using their data and confirmed models to perform optimization on the responses.

3.1. Variables

x_{CaO}, x_{MgO}, x_{CaF_2} and $x_{Al_2O_3}$ represent the respective weight percent of CaO, MgO, CaF_2 and Al_2O_3 in the flux.

3.2. Constraints

The constraints of the model are the lower and upper limits of the flux ingredients. These constraints define the experimental space. From Kanjilal et al[5-8] the lower and upper limits are:

$$15 \le x_{CaO} \le 35 \quad (5)$$

$$10 \le x_{MgO} \le 32.40 \quad (6)$$

$$10 \le x_{CaF_2} \le 40 \quad (7)$$

$$8 \le x_{Al_2O_3} \le 40 \quad (8)$$

There is an additional constraint that the proportions of these ingredients must sum up to 80%. The balance (20%) consists of SiO_2, Fe-Mn, Ni and bentonite all of them with fixed compositions throughout the experiments (Table 1).

$$x_{CaO} + x_{MgO} + x_{CaF_2} + x_{Al_2O_3} = 80 \quad (9)$$

3.3. Objectives

Although our aim here is to demonstrate the feasibility of integrating the MP technique with XVERTD for welding flux optimization, we tried to be realistic in setting the objectives. For instance, Kanjilal et al[6] observed that mechanical properties improved with increase in the amount of acicular ferrite (AF) in the microstructure. This is in agreement with the work of other researchers[9,12,19]. The work of Kanjilal and co-investigators also show that for the particular steel they studied mechanical properties improved with decreasing oxygen content. Generally, maximization of charpy impact toughness is a most desirable mechanical property. Hence we consider the following four single objective cases, namely;
a) The flux designer wants to maximize the acicular ferrite content of the weld-metal;
b) The FD wants to minimize the oxygen content of the weld-metal;
c) The FD wants to maximize the charpy impact toughness of the weld-metal; and
d) The FD wants to maximize Si transfer (Table 2).
The objective functions are the confirmed models from Kanjilal et al.[5-8].

3.3.1. Case 1

The FD wants to determine the levels of welding flux ingredients that will achieve maximum level of acicular ferrite in the weld-metal microstructure. The problem may be stated as:

maximize, $\eta_{AF} = -4.83x_{CaO} + 2.08x_{MgO} - 0.37x_{CaF_2} - 0.69x_{Al_2O_3} + 0.08x_{CaO}x_{MgO} + 0.16x_{CaO}x_{CaF_2} + 0.17x_{CaO}x_{Al_2O_3} - 0.07x_{MgO}x_{CaF_2} - 0.07x_{MgO}x_{Al_2O_3} - 0.01x_{CaF_2}x_{Al_2O_3}$

Subject to:

$$\begin{aligned} & x_{CaO} + x_{MgO} + x_{CaF_2} + x_{Al_2O_3} = 80 \\ & x_{CaO} \ge 15 \\ & x_{CaO} \le 35 \\ & x_{MgO} \ge 15 \\ & x_{MgO} \le 32.40 \\ & x_{CaF_2} \ge 10 \\ & x_{CaF_2} \le 40 \\ & x_{Al_2O_3} \ge 8 \\ & x_{Al_2O_3} \le 40 \end{aligned} \quad (10)$$

For the remaining cases, their respective confirmed models (Table 2) were similarly used as the objective function subject to the same constraints. The constraints will not change because the models were derived under the same experimental conditions.

Table 1. Treatment combination determined by the XVERTD design and results of the experiments.

Sample Nº	Mixture variables composition wt. (%)				Constant composition wt. (%)					Measured responses from experiments			
	CaO	MgO	CaF_2	Al_2O_3	SiO_2	Fe-Mn	Fe-Si	Ni	Bentonite	AF (%)	Impact Toughness at –20 °C (J)	Oxygen (ppm)	Silicon Transfer (ΔSi%)
P1	15.00	15.00	10.00	40.00	10.0	4.0	3.0	1.0	2.0	13	8.8	560	0.207
P2	15.00	15.00	40.00	10.00	10.0	4.0	3.0	1.0	2.0	12	9.8	570	0.065
P3	15.00	32.40	10.00	22.60	10.0	4.0	3.0	1.0	2.0	15	10.5	520	0.140
P4	15.00	17.00	40.00	8.00	10.0	4.0	3.0	1.0	2.0	14	9.8	500	0.027
P5	15.00	32.40	24.60	8.00	10.0	4.0	3.0	1.0	2.0	13	7.8	530	0.108
P6	35.00	15.00	10.00	20.00	10.0	4.0	3.0	1.0	2.0	24	22.2	380	0.092
P7	17.00	15.00	40.00	8.00	10.0	4.0	3.0	1.0	2.0	16	13.7	490	0.126
P8	35.00	15.00	22.00	8.00	10.0	4.0	3.0	1.0	2.0	19	14.4	480	0.064
P9	29.60	32.40	10.00	8.00	10.0	4.0	3.0	1.0	2.0	28	16.7	330	0.132
P10	35.00	27.00	10.00	8.00	10.0	4.0	3.0	1.0	2.0	16	14.7	480	0.043
P11	24.43	23.14	24.43	8.00	10.0	4.0	3.0	1.0	2.0	35	26.0	300	–0.022
P12	15.67	15.67	40.00	8.66	10.0	4.0	3.0	1.0	2.0	26	15.8	350	0.019
P13	25.92	24.36	10.00	19.72	10.0	4.0	3.0	1.0	2.0	28	23.5	320	0.017
P14	23.40	15.00	24.40	17.20	10.0	4.0	3.0	1.0	2.0	36	25.5	300	0.112
P15	19.87	32.40	14.86	12.87	10.0	4.0	3.0	1.0	2.0	35	24.1	320	0.228
P16	15.00	22.36	24.92	17.72	10.0	4.0	3.0	1.0	2.0	10	9.1	600	0.060
P17	35.00	19.00	14.00	12.00	10.0	4.0	3.0	1.0	2.0	20	14.2	470	0.139
P18	22.67	21.63	21.63	14.07	10.0	4.0	3.0	1.0	2.0	16	11.6	540	0.026

Source: Kanjilal et al.[5-8]

Table 2. The Objectives of the flux designer and the corresponding models.

Objective of FD	Objective function to be optimized
Maximize acicular ferrite content	$\eta_{AF} = -4.83x_{CaO} + 2.08x_{MgO} - 0.37x_{CaF_2} - 0.69_{Al_2O_3} + 0.08x_{CaO}x_{MgO} + 0.16x_{CaO}x_{CaF_2} + 0.17x_{CaO}x_{Al_2O_3} - 0.07x_{MgO}x_{CaF_2} - 0.07x_{MgO}x_{Al_2O_3} - 0.01x_{CaF_2}x_{Al_2O_3}$
Minimize oxygen content	$\eta_{O_2} = 63.305x_{CaO} - 12.42x_{MgO} + 6.457x_{CaF_2} + 16.775x_{Al_2O_3} - 0.945x_{CaO}x_{MgO} - 1.557x_{CaO}x_{CaF_2} - 2.061x_{CaO}x_{Al_2O_3} + 0.835x_{MgO}x_{CaF_2} + 0.767x_{MgO}x_{Al_2O_3} + 0.378x_{CaF_2}x_{Al_2O_3}$
Maximize impact toughness	$\eta_{Toughness} = -3.31038x_{CaO} + 0.62389x_{MgO} - 0.26209x_{CaF_2} - 0.84441x_{Al_2O_3} + 0.06680x_{CaO}x_{MgO} + 0.10098x_{CaO}x_{CaF_2} + 0.12913x_{CaO}x_{Al_2O_3} - 0.03063x_{MgO}x_{CaF_2} - 0.02394x_{MgO}x_{Al_2O_3} - 0.00737x_{CaF_2}x_{Al_2O_3}$
Maximize silicon transfer	$\eta_{Si\ Transfer} = 0.012176x_{CaO} + 0.055635x_{MgO} + 0.006303x_{CaF_2} + 0.013559x_{Al_2O_3} - 0.001364_{CaO}x_{MgO} - 0.000063x_{CaO}x_{CaF_2} - 0.000190x_{CaO}x_{Al_2O_3} - 0.001332x_{MgO}x_{CaF_2} - 0.001429x_{MgO}x_{Al_2O_3} + 0.000220x_{CaF_2}x_{Al_2O_3}$

Source: Kanjilal et al.[5-8]

4. Results of the Model

The models were solved using the Lingo 11 software. The results of the MP models for single objective flux formulation situation are presented in Table 3. A comparison of the MP models results with the results of the experiments show that the MP technique is a useful tool for the identification of the optimum formulation for a given response. For instance, the maximum AF percent in the microstucture from the experiments of Kanjilal et al.[6] was 36% (Tables 1 and 3). If decision were based on the result from experiment alone, the formulation that gave 36% AF will be selected as the optimum. We can see from Table 3 that such a decision will be misleading. With the MP optimization approach, a formulation that will give as much as 51.2% AF was identified within the same experimental domain. Optimum formulation from the MP model is 42% higher than that of the experimental value. This is the situation with oxygen content and charpy impact toughness with 17 and 11.4% respective improvement over the results from experiment. The results show that flux formulation based on experiment alone can not be guaranteed to be the optimum formulation. Even in situations where the optimum value from experimental data coincides with the real optimum, there is no quantitative means (optimality criteria) of identifying it. For instance, in the case of silicon transfer (Table 3), where the difference between the experimental result and MP model result is not much (1.3%), there was no means of identifying it as the real optimum because of the lack of mathematical test for optimality.

Although the extreme vertices design (XVERTD) is a proven design of experiment (DoE) method for mixtures and the data from the experiment can be used to develop regression models, relying on

Table 3. Comparison of single objective MP model results with experimental results.

Response	MP Model Results					OptimumValue From Experiment	Difference Between MP and Experiment (%)
	CaO (%)	MgO (%)	CaF_2 (%)	Al_2O_3 (%)	Optimum Value (MP Model)		
Maximize AF	25.05	15.00	31.95	8.00	51.2%	36.0%	42%
Minimize Oxygen Content	27.14	15.00	10.00	27.86	249.2 ppm	300.0 ppm	–17%
Maximize Charpy Impact Toughness	27.42	15.00	10.00	27.58	29.0 J	26.0 J	11.4%
Maximize SiliconTransfer	15.00	15.00	16.15	33.85	0.231%	0.228%	1.38%

the experiments alone to take decision can be misleading as has been seen in this study. To locate the optimum point(s) in the experimental space, it is better to couple it with optimization techniques. The FD can also know from the results of the MP model either it is feasible or not to achieve a desired performance level with the flux ingredients under the prevailing conditions. For instance, it is not feasible to achieve AF content above 51.2%, oxygen content below 249 ppm, charpy impart toughness above 29 J and silicon transfer above 0.231% (Table 3) with the present flux ingredients. If the FD desires weld-metal AF content above 51.2% or oxygen content below 249 ppm for example, then he has to think of changing the flux ingredients, add ferroalloys, or any other action necessary for him to achieve his desired response value(s). In the case of experimental optimization the FD can not know the feasibility or otherwise of achieving his desired response values until after a lengthy and expensive trial and error experiments. For the case under study, it was possible to establish optimality and feasibility with only 18 experiments and as a consequence, it is possible to reduce lead-time and costs associated with extensive experimental weld production and testing.

5. Conclusion

Mathematical programming optimization technique was integrated with XVERTD method for welding flux formulation. The major conclusions are;

- It is feasible to integrate MP optimization technique with XVERTD for the purpose of determining the flux ingredient levels that optimizes desired responses;
- The random character of flux designed through experimental optimization is eliminated because the integration of MP with XVERTD guarantees optimum flux formulation;
- Feasibility or otherwise of achieving the required performance level can be known early with few experiments unlike the case of experimental optimization where feasibility or otherwise is difficult to be ascertained until after a lengthy trial and error experiments; and
- The lead-time, costs of labour, energy and materials usually expended on extensive trial and error experiments can be drastically reduced with the integration of XVERTD with MP optimization techniques in welding flux design.

The MP optimization techniques are not limited to single response optimization situations. MP techniques exist for multiresponse optimization too.

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