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**AN INNOVATIVE CONTROL SYSTEM  
FOR THE GAS NITRIDING PROCESS INCLUDING  
AN INTERACTIVE MODULE TO PREDICT  
THE RESULTS OF THE PROCESS**

**Key words**

Gas nitriding, mathematical model, control system.

**Abstract**

The article presents the concept of a system to control the gas nitriding process containing an interactive module for predicting the results of the process on the basis of mathematical model of growth kinetics of the nitrated layer. The concept of the system is based on comparing the current values of process parameters: temperature, the composition of nitriding atmosphere, and the current value of the nitrogen potential (which is determined based on the signal from the hydrogen sensor) with a pre-established algorithm of changes in process parameters. According to the developed concept, if there are differences, the control system selects one of two options:

- Changing the flow rate of the components of a nitriding atmosphere, in the range of possible changes for a given stand to nitriding, to achieve a predetermined potential value, or
- Setting a new algorithm for changes in temperature and nitrogen potential.

Selecting one of these options by the system will depend on obtaining the slightest difference between on-line calculated result of a process on the basis of mathematical model (for both options) and the assumed result, i.e. the nitrided layer thickness and profile of hardness change.

## **1. Introduction**

The issues of effective design of the processes of the formation of material properties in the direction of multifunctional properties constitute one of the most modern world trends in the area of materials research. For this purpose, mathematical apparatus is being used more frequently and more widely and analytical and numerical mathematical models are being included together with methods of computer intelligence. These tools serve to support the design of surface treatment processes; among these, the gas nitriding process is of a great significance. In the recent years, it has been distinguished by a dynamically growing range of applications, mainly in the automotive and military industries, as well as in the civil aircraft industry. At the same time, requirements are increasing, concerning the specific characteristics of mechanical, chemical, and operational nitrided machine parts and tools. These growing requirements, in combination with an absolute condition of obtaining in a repeatable manner a complex layer structure, have served to stimulate research aimed at widening the spectrum of available process control methods [1–12]. This includes the tests covered by the present paper and concerning the development of methods to assist the process planning and development of control systems. In particular, the article presents the concept of an operating system to the control of gas nitriding process containing interactive module for predicting the results of the process on the basis of a mathematical model of the growth kinetics of nitrided layer wherein the input parameters are the current values of the process parameters: temperature, the composition of nitriding atmosphere, and the current value of the nitriding potential.

## **2. Control System**

### **2.1. General concept**

Regarding the nitriding process control systems used at present, attempts have been made to obtain increasingly precise methods to determine the composition of the nitriding atmosphere. The sensors used in the latest solutions

of automatic systems make it possible to determine the most representative quantity which has an impact on the course of the creation of the layer, i.e., the nitriding potential ( $K_N = P_{NH_3} / P_{H_2}^{3/2}$ ) whose algorithm of changes in the

function of the process time and temperature constitutes the point of reference for the control system. However, a selection of a suitable algorithm of the changes of the nitriding potential and the temperature as well as an application of increasingly better sensors of the nitriding atmosphere composition and temperature does not always guarantee that the expected result is achieved. For various practical reasons, the process may not run as expected. In such cases, we learn that layer structure is different from the expected one only after the completion of the process.

The concept of the system is based on comparing the current values of process parameters: temperature, the composition of nitriding atmosphere, and the current value of the nitrogen potential with a pre-established algorithm of changes in process parameters. According to this, if there are differences, the control system selects one of two options:

- Changing the flow rate of components of a nitriding atmosphere, in the range of possible changes for a given stand to nitriding, to achieve a pre-established potential value, or
- Setting a new algorithm for changes in temperature and nitrogen potential.

Selecting one of these options by the system will depend on obtaining the slightest difference between the on-line calculated result of a process (for both options) and the assumed result, i.e. the nitrified layer thickness and hardness profile.

## 2.2. Mathematical model

The course of the kinetics of the process depends on the speed of particular partial stages. An equation describing the kinetics of its slowest stage is an equation that describes the kinetics of the whole process. The speed of particular stages of the creation and growth of the nitrified layer changes together with the time of the process. A chemical reaction of ammonia dissociation on the surface of the nitrified element is the slowest stage in the initial phase of the layer creation. While the layer is growing, the diffusion of nitrogen in steel is becoming the slowest stage. This results in the fact that the kinetics of the whole process is a complex time function. However, the transport of nitrogen into the steel [13], which is the primary aim of the whole process, is accomplished through the proper selection of the following parameters: temperature, the intensity of the flow of the nitriding atmosphere, and its composition. Experimental data collected for such cases concerning the growth

of particular mono-phase zones in the nitrided layer [14], as well as the growth of such zones in other diffusion systems, e.g., metal-metal [15], indicate a parabolic law of the growth of phase zones:

$$\Delta x_i = k_i \sqrt{t} \quad (1)$$

where

- $\Delta x_i$  – thickness of  $i^{\text{th}}$  phase in  $n$ - phase layer after process time  $t$  (for nitrided layer, the maximum value  $n = 3$ ),
- $k_i$  – kinetic parameter of the growth of  $i^{\text{th}}$  phase, the so-called constant of parabolic growth of  $i^{\text{th}}$  phase,
- $t$  – process time.

Equation (1), with experimentally designated constant  $k_i$ , serves to describe the growth kinetics of phase zones at a given temperature. For practical reasons, it is sufficient in most cases to have knowledge of  $k_i$  value at different temperatures for all the phases of the diffusing system, which makes it possible, in accordance with equation (1), to determine the change of the thickness of a given  $\Delta x_i$  phase in the function of process time. However, this is an oversimplified description of the growth kinetics, which makes it impossible to determine, in the case of a nitrided layer, e.g., nitrogen diffusion coefficients in an individual phase or nitrogen concentrations on phase borders. One cannot foresee, based on this equation, the influence of the remaining phases creating the layer upon the speed of the growth of a given phase. For this reason, it became necessary to develop general mathematical equations describing the dependencies between growth parameters ( $k_i$ ) and diffusion parameters, which facilitated the determination of diffusion coefficients in mono-phase layer zones, as well as the forecasting of the phase growth in the function of the process time. In the developed model, whose particulars were given in paper [9], the final results are the equations given below (2), which, in a direct manner, connect the kinetic parameter of a given phase  $k_i$  with the difference of concentrations on the phase borders and an effective diffusion coefficient in the phase:

$$\sum_{j=1}^n \beta_{i,j} k_i k_j = (D_N^i)_{ef} \Delta c_i \quad (2)$$

where

$$\beta_{i,j} = \left\{ \begin{array}{l} \bar{c}_i \\ \frac{1}{4} (3\bar{c}_i + c_{i,i+1}), j+1 \\ \bar{c}_j \end{array} \right\}$$

$k_i, k_j$  – kinetic parameters of the growth of  $i^{\text{th}}$  and  $j^{\text{th}}$  phase,  
 $\Delta c_i$  – difference of concentrations of the diffusing element on the border  
of  $i^{\text{th}}$  phase.

This is a system of non-linear equations, which can be solved with, e.g., a method of successive approximations (iterations):

$$k_i = \frac{2(D_N^i)_{ef} \Delta c_i}{\sum_{j=1}^n \beta_{ij} k_j} \quad i=1, 2, 3 \quad 1-\varepsilon, 2-\gamma', 3-\alpha.$$

In the method of successive approximations, e.g., for zone  $\varepsilon$ , the following dependencies are obtained:

$$k_\varepsilon^{(1)} = \frac{2(D_N^i)_{ef} \Delta c_\varepsilon}{\sum_{j=1}^3 \beta_{ij} k_j^{(0)}} \quad k_\varepsilon^{(2)} = \frac{2(D_N^i)_{ef} \Delta c_\varepsilon}{\sum_{j=1}^3 \beta_{ij} k_j^{(1)}}.$$

From the obtained system of equations, it is evident that the growth parameters are greater with the greater difference of the concentrations of the borders of phases  $\Delta c_i$ , in accordance with the diagram of the phase equilibrium, which corresponds to the greater effective diffusion coefficient in a given phase –  $(D_N^i)_{ef}$ .

In the case of alloy ferrite, which includes alloy elements with a smaller enthalpy of the creation of nitrides than iron, an important objective for mathematical modelling is (apart from formulae describing a change of the layer thickness), the determination of the profiles of nitrogen dissolved in the set of matrix and nitrogen bound in nitrides of alloy elements. Both of these profiles form the distribution of hardness values in the nitrated layer. As it is well known, alloy elements that form nitrides are characterized by a different chemical affinity to nitrogen. If there is a strong affinity between these elements (Ti, V and Cr above 2.5 wt.-%) and nitrogen, then the front of the reaction of the creation of nitrides is ahead of the front of nitrogen diffusion [9, 10, 16, 17]. Consequently, a sharp inter-phase border is created between the layer and the core. In this case, the analysis of the kinetics of the growth of the diffusion layer is limited to calculations of the growth of its thickness, while making use of the model developed for the processes of internal oxidation [18–20]. In this model, the diffusion of alloy elements is neglected, and it is assumed that there is

a constant concentration of nitrogen on the surface, which is in equilibrium with the nitriding atmosphere. With these assumptions, the thickness of the diffusion layer may be described with the following relationship:

$$\xi^2 = \frac{2}{r} \frac{[c]}{[X]} D_N^\alpha t \quad (3)$$

where

- $\xi$  – thickness of diffusion layer,
- $[c]$  – surface concentration of nitrogen in at. %,
- $[X]$  – concentration of alloy element in at. %,
- $r$  – ratio of number of nitrogen atoms to the atoms of alloy element in nitride,
- $D_N^\alpha$  – nitrogen diffusion coefficient in ferrite,
- $t$  – process time.

In the presence of alloy elements, which are characterized by a less chemical affinity to nitrogen (e.g., Mn, Mo, Cr<2.5 wt.-%) as compared to those mentioned above, the inter-phase boundary the layer–core becomes less sharp [21]. This is due to the advance of the front of the reaction of the creation of nitrides by the nitrogen diffusion front. This excludes the possibility of the application of the abovementioned model. In such cases, the growth kinetics for the diffusion layer is determined by the calculation of the profiles of nitrogen dissolved in alloy ferrite and nitrogen bound in the nitrides of nitride-creating elements. These profiles are obtained by solving standard Fick's laws, which include additionally terms that determine the order and invertibility of the reaction of a new phase creation. With the assumption that the concentration of the diffusing element (nitrogen) on the metal surface is constant and is in equilibrium with the surrounding medium, and while assuming an independence of the nitrogen diffusion coefficient from its concentration, equations to describe the nitrogen concentration change and the change of the substitutional alloy element concentration, with which nitrogen reacts, can be presented in the following form [22]:

$$\frac{\partial c_1}{\partial t} = D_1 \frac{\partial^2 c_1}{\partial x^2} - kc_1 c_2 \quad (4)$$

$$\frac{\partial c_2}{\partial t} = -kc_1 c_2 \quad (6)$$

With the solution of the abovementioned system of equations, it is assumed that a local thermodynamic balance is obtained very quickly. Accepting a solution with the use of Crank-Nicholson's method [23], at each time-step, the profile of nitrogen bound in the nitrides of alloy elements is calculated (with a determined affinity of alloy elements to nitrogen being taken into account), as well as the profile of nitrogen dissolved in alloy ferrite, which takes part in further diffusion and the creation of new nitrides of alloy elements.

### 3. Experimental procedures

#### 3.1. Test stand for nitriding

The nitriding processes were conducted in an industrial furnace equipped with a hydrogen sensor for determining the values of nitrogen potential. The control system set the flow rate for nitrogen, ammonia, and dissociated ammonia according to the preset program, and it is capable of maintaining the preset values of  $K_n$  within certain limits as desired for each process stage by continuous self-correction.

#### 3.2. Nitriding parts

Nitriding parts were made from X38CrMoV5-1 (1.2343, WCL) steel. The nominal compositions of this steel and its heat treatment prior to nitriding are summarised in Table 1.

Table 1. Chemical composition of the materials investigated (in wt-%) and heat treatment prior to nitriding

C	Mn	Si	P	S	Cr	Ni	Mo	W	V	Co	Cu
0.35	0.35	1.00	0.03	0.03	5.0	0.35	0.35	0.3	0.4	0.3	0.3
Hardening: 1030°C. Tempering: 550°C											

### 4. Results

#### 4.1. The choice of process parameters

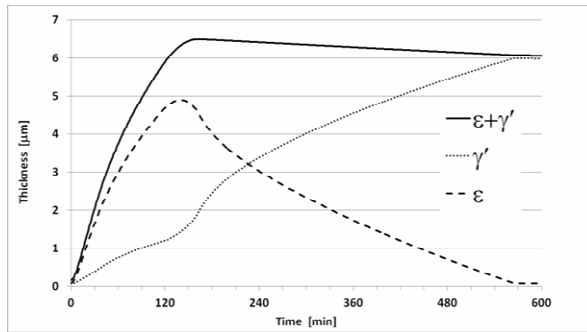
Assumptions concerning the structure of nitrided layer on X38CrMoV5-1 steel:

- Thickness of compound layer – 5–10  $\mu\text{m}$ ,
- Effective thickness of diffusion layer – 150–180  $\mu\text{m}$ .

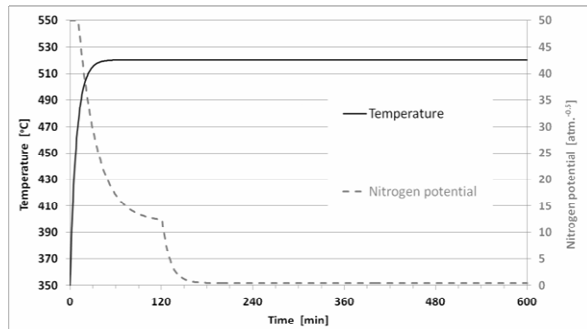
Based on a simulation of the layer growth and based on the database, a two-stage nitriding process was selected (Fig. 1b). Operating parameters were as follows: (Stage 1) 530°C and set-point  $K_n = 15.0 \text{ (atm)}^{-1/2}$ ; (Stage 2) 530°C and set-point  $K_n = 0.38 \text{ (atm)}^{-1/2}$ . The compound layer should reach a thickness

of 5–10  $\mu\text{m}$  for these process parameters in first stage of the process (Fig. 1a). In the second stage of the process, the thickness of the compound layer should not continue to increase, and only the thickness of the diffusion layer should increase.

a)



b)



c)

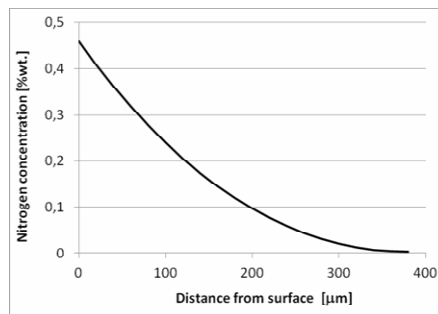


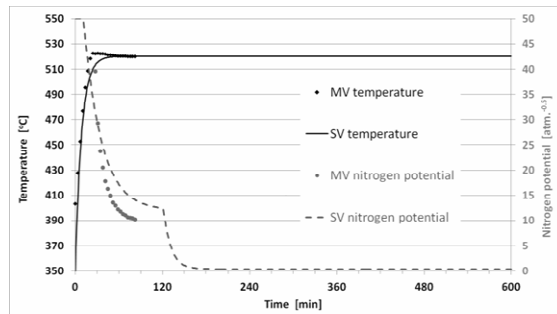
Fig. 1. The simulation of growth kinetics of  $\epsilon$  and  $\gamma'$  zones in compound layer – a); the algorithm changes of temperature and nitriding potential chosen on base of simulation – b); profile of distribution of nitrogen in diffusion layer – c)



#### 4.2. The control of the process run

There were differences between the predetermined algorithm of changes of the nitriding potential and the actual potential changes of nitrogen in the process defined by signal from hydrogen sensor. The control system in a range of possible changes in the flow of ammonia had not led to changes in line with the adopted course. The values of nitriding potential in the first stage of the process were less than assumed (Fig. 2a). As a result, the predicted result of the process, i.e. the thickness of the compound layer after the first stage of the process has not been reached. There were differences between the calculated on-line thickness of the layer and the predicted thickness (Fig. 2b). In this situation, the control system extended the duration of the first stage of the process until a predetermined thickness of the compound layer was reached.

a)



b)

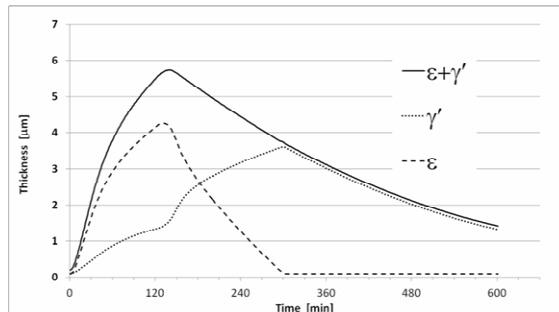
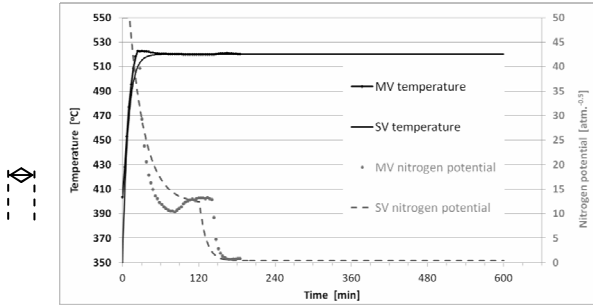


Fig. 2. The differences between the predetermined algorithm changes of the nitriding potential and the actual changes of nitriding potential in the process defined by the signal from hydrogen sensor. MV – values of temperature and nitriding potential measured during process time. SV – setpoint values defined on base of simulation (Fig. 1) – a); the predicted thickness of monophasic zones of the compound layer according to the real value of the nitriding potential after 1.5 h forcing a reaction of control system – b)

a)



b)

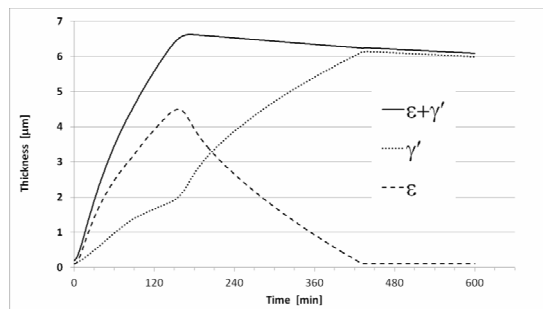


Fig. 3. The correction of the process parameters: the increase of temperature and time prolongation of the first stage of the process – a); the predicted of thickness of the layer according to corrected values of temperature and nitriding potential after 2.5 h – b)

## 5. Discussion

According to the idea of the system, a mathematical model is used for two purposes. Firstly, it provides a simulation of the kinetics of the layer growth, and thus allows determining the most preferred algorithm changes of the process parameters in order to assume the structure of the nitrided layer, i.e. the thickness (carbo) nitrides of iron and concentration profile of nitrogen in the diffusion layer. Secondly, the current parameters determined during the process, i.e. temperature and nitrogen potential are the input parameters of the model, allowing the determination of the current structure layer. In processes where there is a difference between the predetermined algorithm changes of nitrogen potential and automatically delineated, the system corrects the difference by changing the flow rates of components of the nitriding atmosphere. However, in cases of divergence, which are common in industrial practice, is not possible to correct. Then the system automatically based on simulations sets a new algorithm for the change of the potential of nitrogen. An example of this process is presented in this article. In the first stage of the process, nitrogen potential has not reached a predetermined value. This would result, in accordance with the

calculated model of further growth of the layer, in a significant inhibition of growth of  $\epsilon$  phase zone in layer of (carbo) nitrides and, consequently, an inhibition of diffusion zone growth. In this situation, the system has set a new algorithm for potential changes of nitrogen in order to achieve the assumed characteristics of the layer.

## Conclusion

1. A concept of a system for the control of gas nitriding process containing an interactive module of predicting the results of the process on basis of a mathematical model of growth kinetics of a nitrided layer has been presented.
2. The control system allows determining during the process a new algorithm that changes the parameters of the process in order to achieve the assumed characteristics of the nitrided layer. When it is not possible to correct the discrepancies between the actual process and the assumed model the actual algorithm changes the parameters.
3. The proposed concept of the control system of the gas nitriding process expands the possibilities of obtaining reproducible characteristics of nitrided layers.

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## **Innowacyjny układ do sterowania procesem azotowania gazowego zawierający moduł interaktywnego prognozowania rezultatu procesu**

### **Słowa kluczowe**

Azotowanie gazowe, model wzrostu warstwy azotowanej, sterowanie procesem.

### **Streszczenie**

W artykule zamieszczono koncepcję funkcjonowania układu do sterowania procesem azotowania gazowego zawierającego moduł interaktywnego prognozowania rezultatu procesu na podstawie kinetyki wzrostu warstwy azotowanej. Idea działania układu polega na porównywaniu bieżących/aktualnych wartości parametrów procesu temperatury, składu atmosfery i bieżącej wartości potencjału azotowego (wyznaczonego na podstawie sygnału z sondy wodorowej) z założonym wstępnie algorytmem zmian parametrów procesu.

Zgodnie z opracowaną koncepcją, w przypadku zaistnienia rozbieżności układ sterowania wybiera jedną z dwóch opcji:

- zmienia natężenie przepływu składników atmosfery azotującej w celu osiągnięcia założonej wartości potencjału bądź
- wyznacza nowy algorytm zmian temperatury i potencjału azotowego.

Wybór jednej z tych opcji przez układ będzie zależał od uzyskania najmniejszej różnicy pomiędzy na bieżąco prognozowanym rezultatem procesu (dla obu opcji) a rezultatem założonym, tj. grubością warstwy azotowanej oraz profilem twardości.

Dodatkowo układ będzie zawierał moduł wizualizacji wzrostu grubości stref warstwy azotowanej oraz wizualizację zmian profili twardości w strefie dyfuzyjnej warstwy azotowanej w trakcie trwania procesu.

