INFLUENCE OF GAS NITRIDING ON FATIGUE PERFORMANCE OF AUSTENITIC STAINLESS STEEL TYPE AISI 304: PARAMETRIC STUDY AND OPTIMIZATION

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الملخص

يهدف هذا البحث إلى دراسة تأثير عملية النتردة باستخدام غاز الأمونيا في مدى درجة حرارة 400-600 °م ومدى زمن نتردة من 10-50 ساعة ومدى معدل تدفق غاز الأمونيا من 100-600 لتر/ ساعة على مقاومة الكلل لمادة الحديد الأوستنيتي المقاوم للصدأ نوع 304. وهذا النوع من الحديد المقاوم للصدأ له تطبيقات عديدة وهامة في الصناعات الغذائية والتقنية الطبية وبالتالي فإنه دائما مجال بحث للتحسين و التطوير بالطرق المختلفة. الدراسات السابقة حول هذا الموضوع تمت باستعمال الطريقة الكلاسيكية للدراسة وذلك بتغيير عامل واحد كلّ مرة وتثبيت العوامل الباقية. تتطلب هذه الطريقة القديمة الكثير من العينات والعمل التجريبي الذي يعد عالي التكلفة علاوة على استهلاك وقت أطول. إضافة إلى أن الطريقة الكلاسيكية ليست قادرة على اكتشاف التأثير المشترك الذي يحدث عند تغيير أكثر من عامل واحد في نفس الوقت ، كما أنها لا تستطيع أداء عملية مفاضلة للحصول على امثل شروط النتردة التي تعطي أعلى مقاومة للكلل. العوائق السابقة أمكن معالجتها باستعمال علم ومنهجية تصميم التجارب بالطرق الإحصائية. وفقا لذلك، فقد تم تصميم تجربة لدراسة تأثير عملية النتردة على مقاومة الكلل بالطرق الإحصائية. وتم القيام بالتجارب المطلوبة وفق التصميم المعد ومن خلال تحليل النتائج تم تحديد افضل شروط للنتردة والتي تعطي أعلى مقاومة للكلل. هذه الشروط هي درجة حرارة 540 °م و 40 ساعة نتردة ومعدل تدفق غاز الأمونيا 400 لتر/ ساعة والتي أعطت مقاومة كلل قصوى مقدارها 318 ميجابسكال. هذه القيمة تمثل زيادة قدرها 27% مقارنة بالقيمة قبل النتردة وهي 250 ميجابسكال. وبناءً على ذلك فإن استخدام طريقة تصميم التجارب أثبتت فعاليتها في هذا المجال وباستخدامها تمت دراسة تأثيرات العوامل المختلفة لعملية النتردة على مقاومة الكلل بشكل مسهب وفعّال وتم أيضا تحديد عوامل النتردة التي تعطي أقصى مقاومة للكلل لهذا المعدن الهام نسبيا.

ABSTRACT

The aim of the present work is to investigate the fatigue performance of an austenitic stainless steel material type AISI 304 after subjecting it to a conventional gas nitriding process throughout temperature range of 400 - 600 ^oC, nitriding time of 10 - 50 hrs and ammonia (NH_3) flow rate of 100 - 600 litre/hr. Previous work on this subject was performed using the classical method of changing one factor at a time. This old method requires many specimens and extensive experimental work which is both costly and time consuming. Furthermore, the classical method is not capable of detecting the interaction effects between the factors and also can not perform experimental optimization. Previous drawbacks are tackled by using the response surface methodology (RSM) in the design of experiments using statistical methods. Accordingly, RSM design of experiment is created; samples were prepared and gas nitrided according to the design of experiment set up. Fatigue tests were performed on an Avery-Denison fatigue testing machine at a stress ratio R = -1. After analyzing the results, it is found that the fatigue strength of the tested specimens has improved to 318 MPa which represents an extent of 27% by gas nitriding as compared with un-nitrided specimens (250 MPa). Optimum settings for the gas nitriding time, flow rate and temperature factors to obtain the maximum fatigue strength were 540 ^oC, 40 hrs, and 400 liter/hr ammonia gas respectively. The nitrided layer depth was examined by light microscope after grinding and polishing then etching using 2% Nital agent (2 ml nitric acid + 98 ml ethanol). Furthermore, it has been shown that RSM can be applied for studying the behaviour of processes depending on several variables and for performing process optimization. This has been performed in an efficient way and without being necessary to carry out a large number of experiments.

KEYWORDS: Fatigue; Gas nitriding; Stainless steel; Ammonia; Response surface; Optimization.

INTRODUCTION

Austenitic stainless steels are attractive materials for various industrial sectors particularly medical and food industries to combat environmental and corrosive attack. However, their inherently poor tribological behaviour (in terms of high friction and low wear and fatigue resistance) has been the main barrier to wider application under corrosion and dynamic wear conditions. Since then much research and development has aimed to combine improvements in wear, corrosion and fatigue properties [1-3].

The heat treatment and surface hardening processes for steel have undergone many changes over the years. Most of the available technologies in the carburizing field have peaked in terms of their application potential. The nitriding processes on the other hand are enjoying a strong comeback with significant advances in processes and equipment. Gas nitriding is a case-hardening process whereby nitrogen is introduced into the surface of a solid ferrous alloy by holding the metal at a suitable temperature in contact with a nitrogenous gas, usually ammonia [4-6].

The nitriding temperature for all steels is between 400 and 650^oC. Principal reasons for nitriding are:

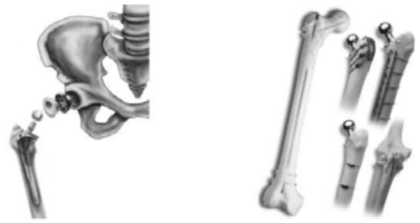
- To obtain high surface hardness.
- To increase wear resistance and antigalling properties.
- To improve fatigue life.

- To improve the corrosion resistance (or keep it while improving other performance parameters).
- To obtain a surface that is resistant to the softening effect of heat at temperatures up to the nitriding temperature.

Nitrided parts are being used in various applications such as in the food-processing, biomedical and automotive industries (see Figure (1)). In food industry, nitrided stainless steel type 302 piston in a liquid-ammonia pump (in Coca Cola factory) lasted for more than five years when replaced a piston made of an un-nitrided 300 series alloy that lasted approximately six months. As well, nitrided type 410 cutting blade showed less wear after completion of five million cuts when used as an alternative of un-nitrided blade which experienced a normal life of only one million cuts. Also, in another application, nitrided type 321, for a motor shaft used in the aeration of orange juice, exhibits very good life that prolonging nine times increasing compared to un-nitrided part [4-6].

In biomedical sector, the austenitic microstructure of austenitic stainless steel is very important due its superior corrosion resistance and nonmagnetic property. This made it compatible for use in magnetic resonance imaging (*MRI*) systems used as diagnostic tools in the medical field [7].

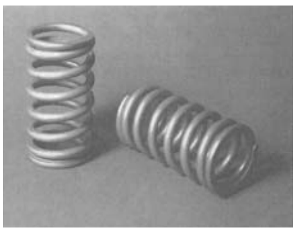
Furthermore, nitriding can be beneficial in other categories as well, such as automotive parts (engine, transmission, chassis, and accessory components), cold-forming tools, and hot-forming tools. Some special applications include screws and cylinders for plastic extrusion, components for rotary internal combustion engines, and synchronizer components for transmissions are ion nitrided to meet close dimensional tolerances, reduction gears for marine steam turbines, deep-drawing punches and hot-forging dies [6].



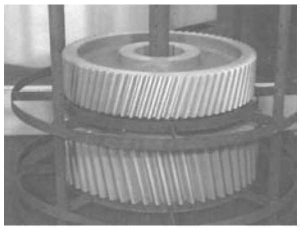
Biomedical (Fracture fixation components)

Valves

Bolts, nuts and washers



Springs



Gears

Figure 1: Nitrided stainless steels parts

Nitriding process can improve fatigue life by producing a hard skin (layer) over a relatively ductile core [1-3]. Furthermore, the additional compressive residual stresses, which are developed on the surface, decreases the likelihood of fatigue failure at that surface. Residual stresses have been produced by means of the volume changes accompanying the nitriding process [4]. For high cycle fatigue (*HCF*), where peak stresses are in the elastic range and the number of cycles required to cause failure is in excess of 10^5, the nucleation of micro-crack in plain specimen constitutes 80 - 90% of the total fatigue life. In low cycle fatigue (*LCF*) where the stresses are high enough to cause macroscopic plastic deformation, fatigue life is correspondingly reduced (usually below 10^5), and the initiation and nucleation of micro-crack in plain specimen may represents only 30 - 40% of the total life [8-10]. In case of nitrided material, the crack initiation usually tends to shift from surface to sub-surface in high cycle fatigue (*HCF*). This may be due to the increased hardness of the surface layer, resulting in better resistance to cyclic slip [5-7]. Several studies were performed on gas nitriding of stainless steel and other steel types as well. A brief review is provided hereafter, which would be treated as a reference for designing the experimental work and for comparing results obtained.

Hussein et. al. [11] investigation showed that nitriding process played the principal role in the improvement of fatigue strength and sub-surface crack nucleation of the maraging steel. Menthe et. al.[12] conducted a series of experiments to study the influence of gas nitriding on the mechanical properties of austenitic stainless steel. His experiments were on the effects of nitriding process on stainless steel type AISI 304L in a temperature range of 375 - 475 ^oC using pulsed-DC plasma with different (N_2 and H_2) gas mixtures and treatment times. He concluded that the treatment influenced the fatigue life, which can be raised by more than 10% at a low stress level (230 MPa). The obtained results showed that plasma nitriding of austenitic stainless steel is a suitable process for improving the mechanical and the tribological properties (especially fatigue strength) without significantly effecting the corrosion resistance of this material.

Bell [1] overviewed the development of low temperature thermo-chemical surface alloying processes. He reported that the fatigue properties of the austenitic stainless steels can be substantially improved by low temperature nitriding. This is mainly due to the formation of a hardened layer which delays the fatigue crack initiation, and the introduction of compressive residual stress which reduces the fatigue crack propagation rate.

Bielawski [13] conducted a nitriding process on chromium steel at a temperature range of 400 - 500 ^oC in ammonia gas atmosphere. The microstructure of the resulted layers was investigated using scanning electron microscopy (SEM) and light microscopy (LM) techniques. Its phase build-up was checked by XRD methods, and the thickness and microhardness of the layers were also measured. He found that, by applying gas nitriding on chromium steel, it is possible to obtain layers with good mechanical properties (microhardness) and good corrosion resistance. Moreover, as a result of gas nitriding process, it was possible to obtain uniform layers during low temperature process. He also found that for nitriding in temperature below 500 ^oC, the obtained layers remained white after etching, which could reflect their good corrosion resistance. All the layers showed very good mechanical properties (high hardness) corresponding to a high nitrogen content in the layers.

In summary, the previous work on gas nitriding process of stainless steel showed that the fatigue properties of the austenitic stainless steels can be significantly improved after nitriding depending upon the treatment conditions. This is mainly due to the formation of a hardened layer which delays the fatigue crack initiation, and the introduction of compressive residual stress which reduces the fatigue crack propagation rate. Also, there is a common conclusion that low-temperature nitriding is preferable to high-temperature nitriding. There is no an overall agreement on the effect of other process conditions such as ammonia flow rate or time of nitriding.

However, previous research work was performed using the classical methods of changing one factor at a time while holding the other factors constant. This methodology requires a lot of specimens and extensive experimental work which is both costly and time consuming. Furthermore, the classical method is not capable of investigating the interaction effects between the factors and also can not be used to perform experiment optimization.

Therefore, the aim of the present work is to investigate the fatigue performance of an austenitic stainless steel material type AISI 304 after subjecting to a conventional gas nitriding process throughout a temperature range of 400 - 600 ^{o}C, a nitriding time of 10 - 50 hrs and ammonia (NH_3) flow rate of 100 - 600 liter/hr. These ranges were estimated based on the previous literature review [1-2] [4-6] [9].

Using the response surface methodology (RSM) in the design of experiment statistical methods will allow studying the effect of the nitriding process in a systematic manner and previous work drawbacks are then tackled. Reason for not using this method in the past is its complex mathematical formulation, which needs a lot of effort and time. These are now facilitated by the recent computer technological development and the generation of powerful statistical packages such as MINTAB program [14].

Principally, conventional gas nitriding is adopted in this research work, because it is cheap and can be used for mass production of industrial parts with all sizes such as gears, valves, bearings etc. Furthermore, due to the affordable technology, this process can be used by small business organizations and private workshops.

MATERIALS AND METHODS

The material used for this investigation was austenitic stainless steel type AISI 304 with a chemical composition shown in Table (1). The material was stress relieved for 3 hrs at 1100 ^{o}C in nitrogen atmosphere, then oil quenched to avoid oxidation. All specimens were subjected to pickling pre-treatment using a hot hydrochloric acid (70 ^{o}C & 50%) to break the oxide film, which is an essential step for gas nitriding process of stainless steels. The response surface methodology (RSM) was employed to determine the required points of experiments (Design of experiment) within considered ranges of nitriding temperature, nitriding time and ammonia flow rate. Anhydrous ammonia gas was used to accomplish the gas nitriding processes. The nitriding processes were conducted using a Pit Furnace type 572 (SIB company) shown in Figure (2). The fatigue specimens shape and dimensions are as shown in Figure (3). The specimens were subjected to gas nitriding process according to experiment matrix obtained from RSM (see Table (2)).

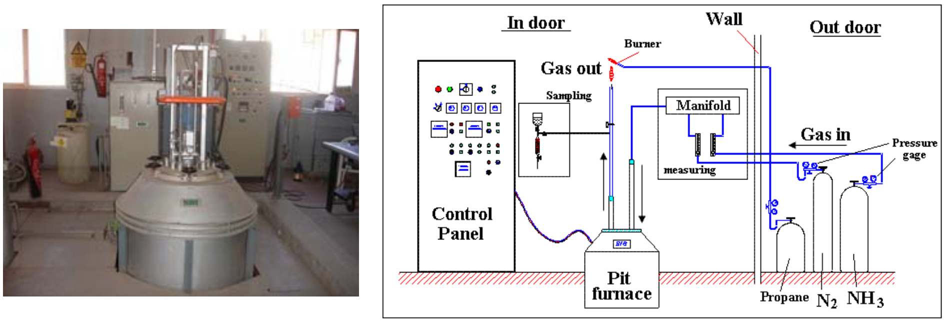


Figure 2: Gas nitriding Pit Furnace model 572 (SIB company)

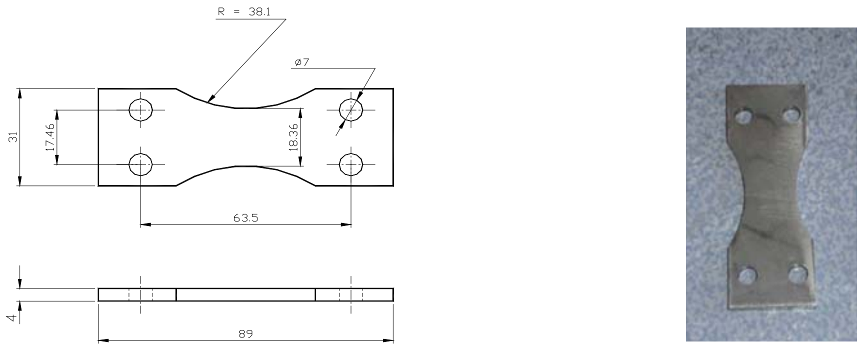


Figure 3: Fatigue test specimen (all dimensions are in millimetres)

Fatigue tests were performed at stress ratio R = -1 (R = σ_{min} / σ_{max} = -1) using Avery-Denison testing machine shown in Figure (4). The pure bending loading condition of the smooth samples is shown in Figure (5). Seven samples were tested for each nitriding process design point and tests were executed up to complete failure of the specimens. The nitrided layer thickness (depth) was examined by light microscope after grinding and polishing then etching using 2% Nital as an etching reagent.

Figure 4: Avery-Denison fatigue testing machine

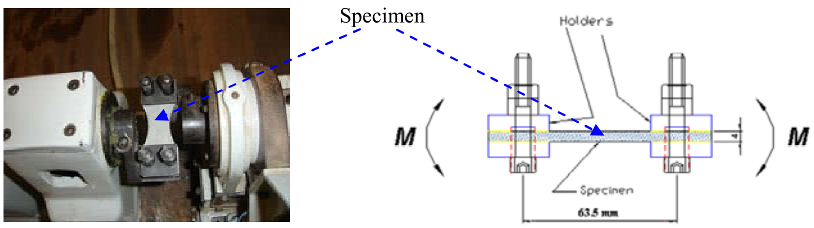


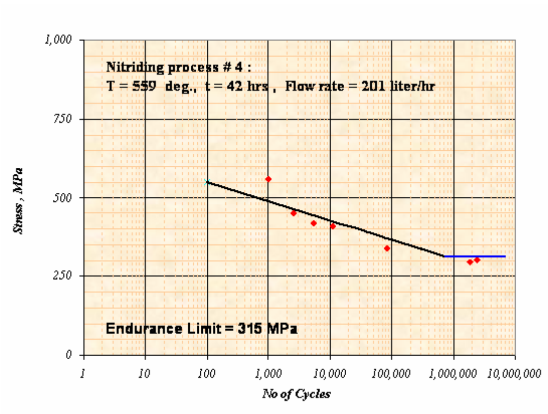
Figure 5: The pure bending loading condition of the smooth samples

RESULTS AND DISCUSSION

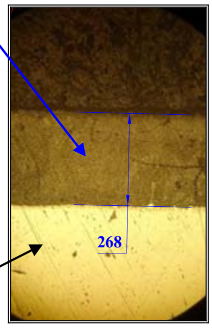
Seven samples for each nitriding process were fatigue tested, and their corresponding fatigue strength (Endurance limit) is obtained after fitting results. Figure (6) shows one of the obtained *S-N* fatigue limit curves corresponding to complete failure and Table (2) summarizes the total experimental results. These results were then analyzed using response surface methodology (RSM), and the interactions of nitriding processing parameters (nitriding temperature, nitriding time and ammonia flow rate) were identified. The effects of nitriding processing parameters on fatigue limit are shown in three dimensional graphs and contours in Figure (7). From Figure (7a), the optimum temperature setting, which is very close to about 540 ^{o}C, can easily be estimated from both the 3D surface plot and the contour plot. This setting will precisely be determined from the optimization chart, which should agree with these plots. Figure (7b) shows that the optimum setting time is between 35 to 40 hrs, which need further clarification from the optimization chart. Furthermore, the optimum setting of the flow rate is very close to about 400 liter/hrs and can easily be observed from both the 3D surface plot and the contour plot. Figure (7c) shows that optimum temperature is further clear when plotted against the flow rat (around 540 ^{o}C). This needs further confirmation from the optimization chart.

Therefore, a third and comprehensive way of presenting these effects is by developing the optimization chart of the fatigue strength with the nitriding conditions which is shown in Figure (8). This Figure shows the optimization chart for the performed fatigue tests on the gas nitrided specimens. The optimization result is shown in the lift column; while the optimum setting of each parameter is shown at meddle of the top row. The behaviour curve of each factor is shown underneath. As shown, an optimum nitriding time is 41.3263 hrs, optimum temperature setting is 540.8755 ^{o}C and optimum ammonia flow rate setting is 396.9666 liter/hr which resulted in fatigue limit of 317.8947 MPa. This achievement represents 27 % increase of the fatigue limit by gas nitriding as compared with the un-nitrided value of 250 MPa. Comparing these results with literature finding, the 525 ^{o}C optimum temperature setting of the gas nitriding process agrees very well with other researcher's findings [11-13]. Furthermore, the optimum 41 hrs nitriding time and the approximately 400 litre/hr ammonia gas flow rate have not been mentioned in the previous work, which is considered as a further contribution.

Finely, it is proved that the response surface methodology (RSM) is a powerful tool for studying the nitriding process parameters effects on fatigue limit, and also to find the optimum nitriding process conditions. Also, the conventional gas nitriding process if properly applied would produce excellent surface properties. This process is suitable for mass production of small and even large mechanical components such as gears, springs and bearings.



(a) *S-N* curve



(b) Surface morphology (200×)

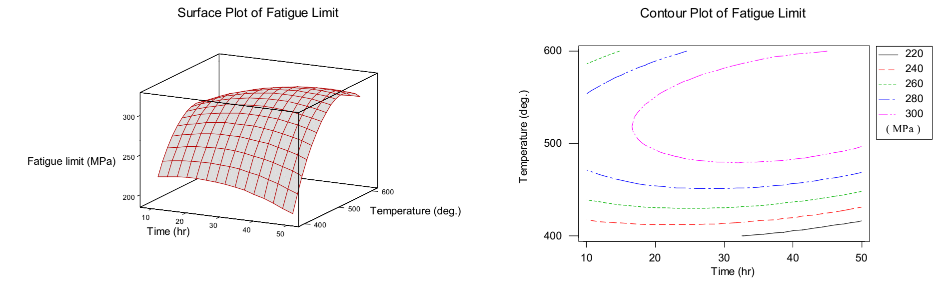
Figure 6: *S-N* curve for fatigue test and layer morphology of nitrided specimen (Design point No 4 (T = 559 ^{o}C, t = 42 hrs , Flow rate = 201 liter/hr)

Table 1: Chemical composition of used AISI 304 austenitic stainless steel

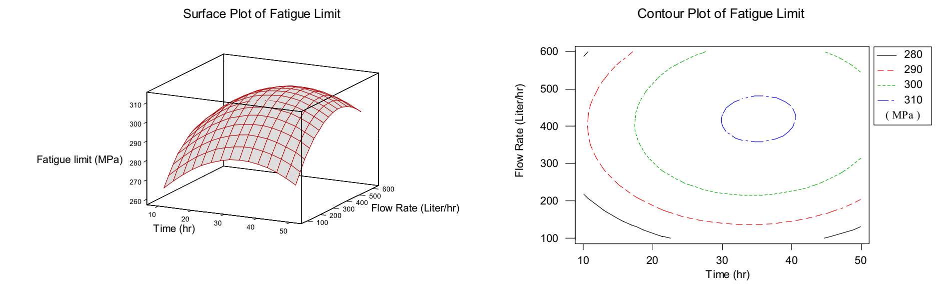
Alloying element	C	Mn	P	S	Si	Cr	Ni
wt.%	0.08	2.0	0.045	0.03	0.75	19	9

Table 2: Gas nitriding process RSM design matrix with results of fatigue strength

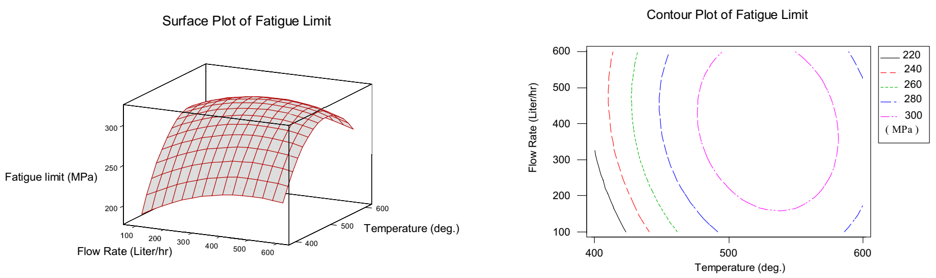
Design point No	Time 10–50 (hrs)	Temp 400-600 (^{o}C)	Flow rate 100– 600 (liter/hr)	Fatigue strength (MPa)
1	18	441	201	250
2	42	441	201	250
3	18	559	201	290
4	42	559	201	315
5	18	441	499	255
6	42	441	499	255
7	18	559	499	280
8	42	559	499	310
9	10	500	350	300
10	50	500	350	300
11	30	400	350	235
12	30	600	350	285
13	30	500	100	275
14	30	500	600	320
15	30	500	350	290
16	30	500	350	325
17	30	500	350	310
Un-nitrided material	---	---	---	250



(a) 3D surface plot and contour for fatigue strength with nitriding time and temperature



(b) 3D surface plot and contour for fatigue strength with nitriding time and ammonia flow rate



(c) 3D plot and contour for fatigue strength with nitriding temperature and flow rate

Figure 7: Effects of nitriding processing parameters on fatigue limit

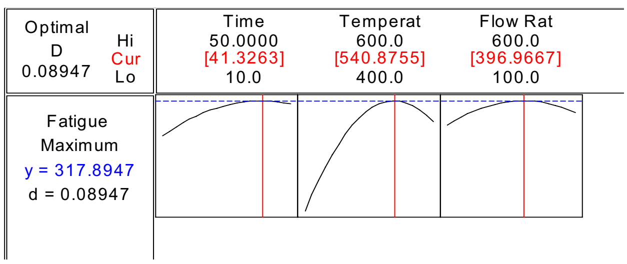


Figure 8: Optimization chart of gas nitriding process for maximum fatigue strength

CONCLUSIONS

From the previous study the following points are concluded:

- A conventional gas nitriding process was successfully performed on a stress relieved austenitic stainless steel type AISI 304 using anhydrous ammonia. The considered nitriding processing parameters were nitriding temperature, nitriding time and ammonia flow rate. Results showed that fatigue strength has improved by 27 % as compared with the un-nitrided case.
- The optimum setting for the nitriding temperature, the nitriding time and ammonia flow rate were obtained.
- The obtained fatigue strength of nitrided specimens were analysed using the response surface methodology, which proved to be a suitable method for comprehensive gas nitriding parametric studies and optimization.

Therefore, in this study, optimum fatigue limit of 318 MPa is obtained by applying a gas nitriding process at 540^{o}C for 40 hrs using 400 liter/hr ammonia flow rate, which represents an increase of 27% as compared with the 250 MPa fatigue strength of the un-nitrided materials. Even though, this percentage represents a good achievement.

ACKNOWLEDGEMENTS

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