

Original Article



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Optimization of DBD plasma reactor parameters for enhanced SO, removal from air: A Taguchi Method

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Abstrac

Background: Sulfur dioxide (SO₂) is an air pollutant that can cause a wide range of health effects. Its' removal can be achieved in one way through oxidation using non-thermal plasma (NTP) technology. This study aimed to optimize the dielectric barrier discharge (DBD) Plasma Reactor Parameters to maximize the SO₂ removal efficiency using the Taguchi Method.

Methods: At first, the operational condition of the DBD plasma reactor was investigated and optimized using a factorial design. Then, the effective parameters on SO_2 removal from air using the selected reactor were optimized using the Taguchi method.

Results: According to the results, the DBD reactor packed with inert ceramic granules (ICGs) was selected as the optimal reactor condition, and analysis of the experiments using the Taguchi method indicated that packing the reactor with ICGs has the most contribution in the SO₂ removal from air. The maximum removal efficiency of the optimized reactor was achieved as 100% at the optimum condition of SO₂ concentration of 600 ppm, flow rate of 2 lpm, and voltage of 25 kV. Also, the result of the validation test showed that the results of the experiments and the model were in good agreement. Conclusion: In this study, following the Taguchi method, the effects of parameters influencing the SO₂ removal using a plasma reactor were analyzed. According to the results, it was concluded that the ICGs can be a good choice for packing the DBD reactor for maximizing the SO₂ removal efficiency, which was increased by 100% at the optimum condition.

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Introduction

Air pollution is one of the greatest environmental risks to health. According to the WHO reports, nine out of ten people breathe polluted air, which kills 7 million people every year. In 2019, 99% of the world's population was living in places where the WHO air quality guidelines were not met. Some 89% of those premature deaths occurred in low- and middle-income countries, and the greatest number in the WHO South-East Asia and Western Pacific Regions. Household combustion devices, motor vehicles, industrial facilities, and forest fires are common sources of air pollution. Pollutants of major public health concern include particulate matter, carbon monoxide, ozone,

nitrogen dioxide, and sulfur dioxide. By reducing air pollution levels, countries can reduce the burden of disease from stroke, heart disease, lung cancer, and both chronic and acute respiratory diseases, including asthma (1).

SO₂ is one of the most important air pollutants, which is released into the air due to the consumption of high-sulfur content fossil fuels from various sources such as industries and heavy vehicles (2). This pollutant causes a wide range of health effects from respiratory discomfort and mucous irritation to genetic abnormalities and death (3,4). The sulfur content of diesel fuels used in developing countries is 30 times more than that recommended by the EPA (15 ppm). During the fuel combustion process,

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almost all of its sulfur content is converted into SO_2 , so it can be concluded that exposure to this pollutant is higher than the permissible exposure limit (5 ppm) in developing countries (5).

Many researchers have investigated different air pollution control technologies for simultaneously controlling the air pollutants using thermal and non-thermal plasma (NTP), catalytic decomposition, physical and chemical absorption, scrubbing, etc. (6-10). One of the main challenges in catalytic conversion of pollutants is the poisoning effect of sulfur oxides on catalysts, which has made the simultaneous control of pollutants difficult (11-13). Therefore, controlling this pollutant is a serious challenge, especially in developing countries that use high-sulfur content fuels.

SO, removal can be achieved in one way by reducing it into elemental sulfur using different reductant catalysts. However, due to the catalysts' poisoning by SO₂, the efforts to obtain the sulfur poisoning-resistant catalyst are still ongoing (14-16). Another way for SO, removal is the oxidation of it, which leads to the formation of sulfuric acid (17,18). Sulfuric acid is formed by the reaction of sulfur trioxide (SO₂) with water. According to the results of related studies, the removal of SO₂ using NTP is mainly through the oxidation process, which leads to the formation of SO₃. This very unstable gas can be used to form sulfuric acid. Regarding the removal of SO, using NTP through the oxidation way, the efficiency of the plasma reactor packed with dielectric granules is much higher than that of the empty plasma reactor (19). Among the dielectric granules, active ceramic granules that contain more than 99% alumina are very good absorbers for SO₂, but they get saturated very quickly, so it is better to use inert ceramic granules (ICGs), which can also be used as a catalyst bed support. These granules don't have any absorption capacity for SO2 and just increase the plasma discharge strength.

This study aimed to investigate the efficiency of an NTP reactor filled with ICGs for SO_2 removal using the Taguchi method and compare it to an empty plasma reactor. Also, before that, it was aimed to investigate the optimal conditions of the plasma reactor for the best discharge formation.

Materials and Methods Experimental Apparatus

The required experimental equipment includes: power supply (Pulsed-DC, Laser and Plasma Research Institute in Shahid Beheshti University, IRAN), continuous gas flow generator system (Researcher-made and nationally patented No. 109021(20), IRAN), plasma reactor, and gas analyzer (MRU (Vario-plus), Germany). The power supply used in this study was an oscillatory Pulsed-DC power supply with variable voltage and frequency. The power supply used in this study was capable of applying

different voltages of 2–25 kV (peak to-peak) by varying the duty cycles of 1-10%. Duty cycle is the pulse active time divided by the total period of a pulse. A continuous gas flow generator system was used to prepare different concentrations of SO_2 gas. A DBD plasma reactor packed with ICGs was used for the removal of SO_2 from air, and its condition was optimized as described below. Gas phase products were analyzed by the real-time analyzer of MRU (Vario-plus) made in Germany, with an accuracy of 10 ppm.

Experimental Design and Analytical Procedure

In the present study, a multilevel categoric factorial design technique (using statistical software of Design-Expert version 11) was used to determine the optimal condition of the reactor in terms of the plasma discharge formation threshold voltage, and the Taguchi method (using Qualitek-4 software) was used to optimize the effective parameters for maximizing SO₂ removal efficiency. Studied parameters for determining the optimal condition for the studied plasma reactor are presented in Table 1. The effective parameters on SO₂ removal efficiency using the NTP reactor include gas concentration (600, 800, and 1000 ppm), gas stream flow rate (2, 2.5, and 3 lpm), and applied voltage of the power supply (13, 18, and 25 kV).

A cylindrical quartz DBD plasma reactor with 30-mm and 15-mm outer diameter and wall thickness, respectively, was chosen as the NTP reactor to study the effects of the intended parameters mentioned above. After optimizing the plasma reactor condition, which its complete information is given in the results section, the internal electrode placed on the axis of the quartz tube was chosen a 22-mm diameter stainless steel rod and aluminum paste attached on the outer surface of the quartz tube acted as a ground electrode for a length of 6 cm. The discharge gap (2.5 mm) between the internal electrode and quartz tube was packed with ICGs with 1.7 to 2 mm diameter. The specifications of the ceramic granules used in this study are given in the Table S1.

Validation Test for the Optimal Condition of the Reactor in Terms of the Plasma Discharge Formation Threshold Voltage

Regarding the validation of the selected model for the optimal condition of the plasma reactor, one test is

Table 1. The studied parameters and their changing levels for optimizing the plasma reactor based on the multilevel categorical design technique

Parameters	Levels	Level 1	Level 2	Level 3
A: External Electrode	3	AL large grid	Al fine grid	AL paste
B: Internal Electrode	3	Rod 22 mm	Rod 21 mm	Rod 16 mm
C: Reactor Packing Condition	3	E*	G**	C***

^{*}E: Empty reactor; **G: Reactor packed with glass granules; ***C: Reactor Packed with ceramic granules.

performed in the optimal conditions determined by the software to check and confirm the agreement between the results obtained from the model and the experiments.

Taguchi Method

To determine the impact of various input parameters on the given response, the proper experimental design is essential. Generally, the removal efficiency of a specific air pollutant is related to its input parameters by nonlinear mathematics. On the other hand, only the lower and upper values of each studied parameter are not sufficient to predict the nonlinear curve. Consequently, three levels have been considered for each factor. A design with all possible combinations of all the input factors is called a full factorial design. For k (= 4) factors, 3 factors at 3 levels and one factor in 2 levels (e.g., this study), a full factorial design has 54 runs. Such a large number of experiments is difficult to carry out. The Taguchi orthogonal array (L9) table provides the design of experiments considering the nonlinear behavior of parameters. So only nine experiments for such factors were chosen as the basis for experimental data generation.

Results

To determine the range of applied voltage to the plasma reactor, firstly, the threshold voltage for plasma discharge formation must be determined for three different conditions of the studied plasma reactor (empty reactor, reactor filled with glass granules, and reactor filled with ceramic granules). Threshold voltage or corresponding duty cycle is the minimum value at which microdischarges become visible (21). For this purpose, the effective parameters on the threshold voltage or corresponding duty cycle, in terms of the type of dielectric granules filling the plasma reactor, the diameter of the inner electrode, which determines the gap of the plasma environment, and the type of the outer electrode, were investigated using a multilevel categoric factorial design technique. For the above three studied parameters, each in three levels (Table 1), 27 experiments were determined using the mentioned design experiment technique, which covers all possible conditions.

Twenty-seven runs determined using a multilevel categoric factorial design are given in the Table S2. The experimental data were analyzed using the design of experiment software to identify the significant parameters. The data were fitted to the appropriate models. The ANOVA results for the selected 2FI model for the intended response are presented in Table 2.

According to ANOVA statistical results for the selected 2FI factorial model (P<0.0001), the indices of R², adjusted R², predicted R², adequate precision, and CV% indices were 0.88, 0.85, 0.79, 17.74, and 25.20, respectively.

Considering that the investigated parameters are qualitative, each in several levels, only the coded model exists. Because the selected model contains more than 12 categorical equations. To obtain the threshold duty cycle using the selected model, in the range defined for the studied variable according to Table 1, the coded values for each variable must be entered into the model. The final model with coded factors is as follows:

Duty Cycle=+4.15+0.4074*A[1] -1.04*A[2]+2.52*B[1]-0.8148 B[2]+2.07*C[1]-1.48*C[2]

The predicted values of the duty cycle resulting from the model against the actual values resulting from the experiments are shown in the Figures S1.

Optimization and Validation of the Threshold Duty Cycle or Equivalent Voltage for Plasma Discharge Formation

To minimize the threshold duty cycle or voltage for plasma discharge formation, the optimal conditions obtained from the software were the ceramic granules as a dielectric material filling the reactor, an inner electrode with a diameter of 22 mm, and an outer electrode made of aluminum fine grid. To confirm the selected model, one test was performed under the mentioned optimal conditions. The results of the validation test are presented in Table 3.

The results of the qualitative investigation of the plasma reactor in terms of the values obtained for the threshold duty cycle, along with the investigated variables, are shown in the Figures S2.

Table 2. The ANOVA results for the selected factorial model

Source	Sum of Squares	df	Mean Square	F-value	P value	
Model	165.56	6	27.59	25.25	< 0.0001	Significant
A: External electrode	14.74	2	7.37	6.75	0.0058	
B: Internal electrode	89.19	2	44.59	40.81	< 0.0001	
C: Dielectric material	61.63	2	30.81	28.20	< 0.0001	

Table 3. The results of the validation test at the optimal condition for the threshold voltage

Pagnanag	Variable			Validation test	Confidence interval of 95%	
Response	Outer electrode	Inner electrode	Dielectric granule type	result	Low	High
Threshold duty cycle	Al fine grid	22 mm Rod	ceramic granules	1	-2.52	2.37

According to this figure, the optimal condition in terms of threshold duty cycle of plasma discharge formation was: ceramic granules as a dielectric material, an inner electrode with an outer diameter of 22 mm, and an outer electrode of Al fine grid.

Investigation of SO₂ Removal Efficiency Using a Plasma Reactor Filled with Ceramic Granules (C) and Comparing it to an Empty Plasma Reactor

This part of the study was conducted to investigate the performance of a plasma reactor filled with ceramic granules and compare it to an empty plasma reactor. Finally, the optimization of the effective parameters on the SO₂ removal in the studied reactors was done by the Taguchi technique. The investigated factors include gas concentration, gas stream flow rate, and applied voltage of the power supply.

At first, to make sure that the studied dielectric granules do not have any absorption effect, the absorption of SO₂ by the dielectric granules was investigated. For this purpose, the concentration of SO₂ gas was stabilized at the desired value just before the reactor, and then, when the plasma was turned off, the diluted gas flow was directed to the reactor, which caused a sudden decrease in the concentration of the diluted gas in the reactor which was due to changing the gas path from before the reactor to the reactor. But after a short time, the concentration of the gas stream passing over the reactor returned to the same value as the stabilized concentration before the reactor, which confirmed the lack of absorption of intended granules.

Then, in the next step, the power supply was turned on, and the applied voltage to the reactor led to the formation of purple discharge and plasma formation. As a result, the concentration of SO_2 gas suddenly decreased, in less than 90 seconds (depending on the gas flow rate), which indicated the maximum efficiency in removing the SO_2 gas. Afterwards, the reduced concentration of SO_2 gas remained constant with a slight fluctuation during the half hour of the investigation period. The experimental results of the SO_2 removal by the DBD plasma reactor in two different conditions of the empty DBD plasma reactor, as E condition, and the DBD plasma reactor packed with ceramic granules as C condition (SO_2 concentration = 600 ppm, 2 L/min, Duty cycle = 8%), are shown in Figure 1.

The residence time of the gas inside the reactor depends on the physical characteristics of the reactor and the gas flow rate. For the studied reactor, the maximum pollutant residence time in the plasma discharge zone is 0.34 seconds, which corresponds to the minimum studied flow rate (2 lpm).

Statistical Analysis

Nine tests defined by Taguchi's method depend on the studied parameters and their studied levels. The Taguchi L9 orthogonal array of the simulations designed and

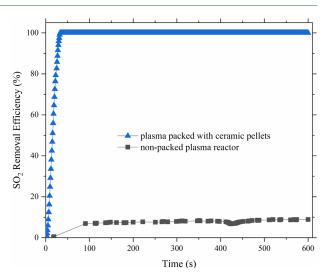


Figure 1. SO_2 removal efficiency of empty DBD plasma reactor and DBD plasma reactor packed with ceramic granules (SO_2 concentration=600 ppm, 2 L/min, Duty cycle=8%)

obtained results are shown in the Table S3.

According to the ANOVA results in Table 4, the plasma reactor type has the most variance, and the voltage is indicated the second place. To check the significance of the studied factors in the Taguchi method, each factor's F-ratio should be greater than 5.3, 9.2, and 29.45 for confidence levels of 90%, 95%, and 99%, respectively.

According to the F-ratio values in Table 4, the confidence level of 90% was chosen, and the insignificant factors (SO₂ concentration and gas flow rate) were pooled. The values of the F-ratio were calculated after pooling these two mentioned factors (Table 5). The percentage contribution of each factor to the SO₂ removal efficiency of the studied plasma reactor performance is shown in Table 5. The percentage contribution of the plasma reactor type was the greatest, approximately 91% (90.924), with that of applied voltage to the plasma reactor being 5.008%.

Level average response analysis can be done based on the average S/N ratio at each level of each factor, and plotting it. In the resulting plots, peak points indicate optimal conditions. Table 6 contains the average effect response for S/N ratios, and the corresponding plots are shown in Figure 2. The results have been analyzed using the Taguchi technique to rank the factors that can affect the SO₂ removal process. Also, all cases between the studied factors are shown in the Table S4.

Confirmation Test

The confirmation test was performed at the optimum level of the studied factors. The confirmation test showed that the SO_2 removal efficiency is 100% and the 90% confidence intervals for the S/N ratio are 40.21.

Discussion

In this study, which was conducted in two steps, the performance of a plasma reactor filled with ceramic

Table 4. ANOVA analysis of S/N ratio

Factor	Degree of freedom (DOF)	Sum of squares (S)	Variance (V)	F-ratio (F)	Pure sum (S')	Percent, P (%)
(A)SO ₂ concentration (ppm)	2	14.962	7.481	1.583	5.511	0.568
(B) Gas flow rate (Lpm)	2	14.719	7.359	1.557	5.267	0.542
(C) Voltage (kV)	2	48.922	24.461	5.176	39.471	4.068
(D) Plasma reactor type	1	886.882	886.882	187.679	882.156	90.924
Other/error	1	4.724	4.724			3.988
Total	8	970.212				

Table 5. Pooled ANOVA analysis of S/N ratio

Factor	Degree of freedom (DOF)	Sum of squares (S)	Variance (V)	F-ratio (F)	Pure sum (S')	Percent, P (%)
(A)SO ₂ concentration (ppm)	(2)	(14.962)		POOLED	(CL=49.35%)	
(B) Gas flow rate (Lpm)	(2)	(14.719)		POOLED	(CL=66.01%)	
(C) Voltage (kV)	2	48.922	24.461	5.176	39.471	4.068
(D) Plasma reactor type	1	886.882	886.882	187.679	882.156	90.924
Other/error	5	31.405	6.881			5.008
Total	8	970.212				100%

Table 6. Average effect response for signal-to-noise ratios

	Factor					
	(A) SO ₂ concentration (ppm)	(B) Gas flow rate (Lpm)	(C) Voltage (kV)	(D) Plasma reactor type		
Level 1	32.894	32.971	27.904	38.19		
Level 2	30.823	30.418	32.415	17.132		
Level 3	29.794	30.121	33.192			
Maximum-minimum	3.099	2.849	5.288	21.057		
Rank	3	4	2	1		

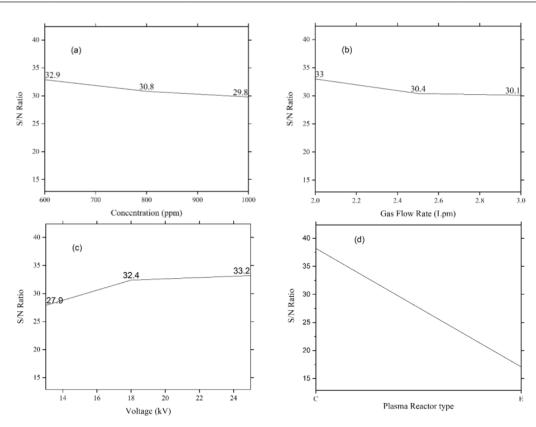


Figure 2. Level average response plots by S/N ratio: (a) SO₂ concentration, (b) Gas flow rate, (c) applied voltage, and (d) plasma reactor type

granules for the removal of SO_2 from air was investigated and compared with an empty plasma reactor. At the first step, the threshold voltage of the plasma reactor was investigated and optimized using a multilevel categoric factorial design technique, and then, the performance of the selected reactor (as optimum reactor condition), which was a plasma reactor filled with ceramic granules, was investigated for the removal of SO_2 from air and its performance was compared with the empty reactor one.

According to the results of the first step (Tables 3 and 4), the effect of the three studied variables on the response of threshold duty cycle or corresponding voltage, and also the selected model with coded terms for this response, was significant (P<0.0001). Considering that the investigated parameters are qualitative, each in several levels, only the coded model exists. The model with coded terms can be used to predict the response for specific levels of each factor. By default, the highest level of each factor is +1 and the lowest level of each factor is -1. The coded model is useful for identifying the relative effect of the factors, which is possible by comparing the coefficients of the factors. Also, the high values of predicted R² and adjusted R² show that the selected model is representative of the system. The optimal value for adequate precision (signalto-noise ratio) is greater than 4, which has been achieved in this study. The value of adequate precision confirms the fit of the model with the data. Also, as can be seen from the figure of predicted duty cycles resulting from the model against the actual values resulting from the experiments, all the points are located around the 45-degree line, which confirms the appropriateness of the model.

Regarding optimization and validation of the threshold duty cycle or equivalent voltage for plasma discharge formation, one test was performed under the mentioned optimal conditions (ceramic granules as a dielectric material filling the reactor, an inner electrode with a diameter of 22 mm, and an outer electrode made of aluminum fine grid). The results presented in Table 3 show that the validation test results lie within a confidence interval 95%, so the validation of the selected threshold duty cycle or equivalent voltage for plasma discharge formation was confirmed.

Regarding qualitative investigation of the plasma reactor in terms of threshold duty cycle, the optimal condition in terms of threshold duty cycle of plasma discharge formation was: ceramic granules as a dielectric material, an inner electrode with an outer diameter of 22 mm, and an outer electrode of Al fine grid. As it is clear in the intended figure in Figures S2, and according to the experimental results, the threshold duty cycle or corresponding voltage for plasma discharge formation in the reactor with the external electrode of Al fine grid is slightly lower compared to the AL paste. Therefore, in real conditions, the outer electrode in two different types of Al fine grid and AL paste was investigated, while the reactor

was filled with ceramic granules, and the inner electrode was a 22 mm Rod. The results of pre-tests showed that, despite the lower plasma threshold voltage using the Al fine grid (as the outer electrode), when the AL paste is used as the outer electrode, the SO₂ removal efficiency is higher. The reason of that the Al fine grid leads to a lower threshold voltage for the plasma discharge formation is the corona formation at the sharp edges of the grid (22). However, the AL paste is completely uniform and does not have sharp edges, which results in uniform plasma discharge formation that leads to a higher SO₂ removal efficiency. Therefore, in real conditions, the outer electrode of the plasma reactor was selected as an AL paste, the inner electrode of a 22 mm 316 steel rod, and the plasma reactor was filled with ceramic granules.

According to the results of plasma threshold voltage optimization in different reactor conditions (empty plasma, plasma filled with glass granules, and plasma filled with ceramic granules), since the threshold voltage of plasma formation in empty plasma reactor (13 kV) is higher than that of the packed plasma reactor, to compare the efficiency of the reactor filled with ceramic granules with an empty reactor, the threshold voltage was selected at 13 kV, which corresponds to duty cycle 6%. The range of investigated variables is the gas concentration of 600, 800, and 1000 ppm, the gas stream flow rate of 2, 2.5, and 3 lpm, and the applied voltage of the power supply of 13, 18, and 25 kV.

After these steps and when the changing levels of studied parameters were determined, and when it was confirmed that the ceramic granules are inert and had no adsorption, the performance of the plasma reactor filled with ceramic granules for the removal of SO, from air was investigated and compared to an empty plasma reactor. According to the results (Figure 1), the performance of the plasma reactor filled with ceramic granules was significantly higher than that of an empty reactor. This may be due to the formation of microdischarges in fine cavities of the porous surface of the ceramic granules, and also may be due to increasing the electrical field in contact points of granules and between granules and electrode surface, which leads to increasing the strength of plasma discharge, which increases the chance of electron-impact reactions responsible for the gas pollutant removal

Comparing the results of this study on the removal of SO_2 by an empty NTP reactor with other similar studies showed that the efficiency of the empty plasma reactor in SO_2 removal is low, depending on the gas concentration (e.g., 10% in 300 ppm) (22,23). This efficiency increases by reducing the initial gas concentration (24), and also by adding some additives such as ammonia (22).

Also, according to the results of statistical analysis using the Taguchi method, it was indicated that the plasma reactor type and then the applied voltage of power supply are most important factors, respectively, and the order of parameters affecting maximum removal efficiency were the plasma reactor type, applied voltage to plasma reactor, SO2 concentration, gas flow rate, and among investigated interaction (Table S4), the SO2 concentration and applied voltage have the maximum interaction. SO2 concentration – gas flow rate and gas flow rate – applied voltage, are another pair where the interaction between them is significant. Also, the experimental results confirmed the validity of the Taguchi technique for optimizing the SO₂ removal efficiency parameters during conducting the confirmation test.

Conclusion

In this study, two optimizations of threshold voltage for plasma discharge formation and SO, removal efficiency of a packed-bed DBD plasma reactor were investigated. Following the Taguchi method, the effects of parameters influencing the threshold voltage for plasma discharge formation, and the SO₂ removal efficiency of the plasma reactor were analyzed. Analysis of the S/N ratio has been applied for finding out the relative contribution and the optimum factor level combination for the maximum SO, removal efficiency. According to the results, the plasma reactor type is the most predominant factor. The importance of the factors on the SO, removal efficiency was ranked in Table 6. The most important parameters, according to their relative significance, are the plasma reactor type and the applied voltage of the power supply, respectively. The maximum SO, removal efficiency was obtained by setting SO, concentration to 600 ppm, gas flow rate to 2 Lpm, the voltage to 25 kV, and the C type of plasma reactor (plasma reactor packed with ceramic granules). It was found that the SO, removal efficiency was increased by 100% at the optimum conditions, which they determined by the Taguchi optimization method.

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Competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

Ethical issues

The Ethical code obtained for the Ph.D. thesis from which this manuscript is derived is IR.TMU.REC.1396.621.

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Supplementary files

Supplementary file 1 contains Tables S1-S4 and Figures S1 and S2.

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