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Babaie, M., Talebizadeh, P., Saha, Suvash C., Rahimzadeh, H., Zare, Firuz, & Brown, Richard J. (2012) Residence time investigation in a co-axial dielectric barrier discharge reactor. In Gu, YuanTong, Saha, Suvash C., & Liu, Gui-Rong (Eds.) *Proceedings of the 4th International Conference on Computational Methods (ICCM2012)*, Crowne Plaza, Gold Coast, Australia.

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Residence Time Investigation in a Co-axial Dielectric Barrier Discharge Reactor

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Abstract

The residence time distribution (RTD) is a crucial parameter when treating engine exhaust emissions with a Dielectric Barrier Discharge (DBD) reactor. In this paper, the residence time of such a reactor is investigated using a finite element based software: COMSOL Multiphysics 4.3.

Non-thermal plasma (NTP) discharge is being introduced as a promising method for pollutant emission reduction. DBD is one of the most advantageous of NTP technologies. In a two cylinder co-axial DBD reactor, tubes are placed between two electrodes and flow passes through the annulars between these barrier tubes.

If the mean residence time increases in a DBD reactor, there will be a corresponding increase in reaction time and consequently, the pollutant removal efficiency can increase. However, pollutant formation can occur during increased mean residence time and so the proportion of fluid that may remain for periods significantly longer than the mean residence time is of great importance.

In this study, first, the residence time distribution is calculated based on the standard reactor used by the authors for ultrafine particle (10-500 nm) removal. Then, different geometries and various inlet velocities are considered. Finally, for selected cases, some roughness elements added inside the reactor and the residence time is calculated. These results will form the basis for a COMSOL plasma and CFD module investigation.

Keywords: Non-thermal plasma, dielectric barrier discharge, Residence time distribution, axisymmetric model

Introduction

Non thermal plasma (NTP) is a promising technology for emission reduction of exhaust gases. Plasma is the fourth state of matter consisting of positive and negative charges which have a tendency to remain overall electrically neutral over large length scales. Vehicle exhaust gases, both diesel and gasoline, undergo chemical changes when exposed to plasma. Various kinds of NTP reactors have been studied by researchers. The majority of researchers have used dielectric barrier discharge (DBD) reactor (Yamamoto, Rajanikanth et al. 2003; Rajanikanth, Subhankar et al. 2004). The DBD reactor has been chosen as the most suitable configuration due to the simplicity and effectiveness for exhaust gas treatment.

Numerical modeling is very useful tool for better understanding of the flow pattern and residence time distribution inside the DBD reactor. The idea of using the distribution of residence times (RTD) in the analysis of chemical reactor performance was first proposed in a pioneering paper by MacMullin and Weber (MacMullin and Weber 1935). However, the concept did not appear to be used extensively until the early 1950s, when Prof. P. V. Danckwerts (Danckwerts 1953) gave organizational structure to the subject of RTD by defining most of the distributions of interest. In a reactor, the various atoms in the feed spend different times inside the reactor. In other words, there is a distribution of residence times of the material within the reactor. The time the atoms have spent in the reactor is called the reactor residence time. In any reactor, the distribution of residence times can significantly affect its performance (Fogler 1999).

In this study a finite element based software called COMSOL Multiphysics has been used to model the RTD. Part of the modeling which has already been completed is the initial step to improving plasma DBD reactor for emission treatment application.

Basic assumptions and regulations

There is a clear symmetry in reactor geometry, therefore a simple 2D axisymmetric model of the reactor was set up and used. This will take whatever geometry you create and rotate it about an axis and it is ideal for problems with symmetry about an axis. The basic assumptions of this reactor are that the flow enters the reactor as fully developed flow and that there are no other effects that need to be considered. The reactor dimensions are as follows:

- Reactor Outer Radius: 11 mm
- Reactor Inner Radius: 10,9 and 8 mm
- Reactor length: 200 mm
- Entry velocity: 2.5 m/s to 25 m/s
- Fluid: Air at 300 K

The first step to in setting of this problem in COMSOL is to choose the type of model. In our problem we selected the 2D axisymmetric option for our model. After that we defined the desired physics. To find the appropriate physics, the Reynold's numbers were calculated. We considered five different velocities in this modeling. These velocities are 0.025 m/s, 0.25 m/s, 2.5 m/s, 5 m/s and 25 m/s. The Reynold's number corresponds to these velocities were 3.46, 34.6, 346, 692 and 3460 respectively. When the flow was laminar ($Re < 2300$), we chose purely laminar flow model and when the flow was turbulent, we used a turbulent model in COMSOL. After that there were two options for selecting the type of simulation in COMSOL: a stationary or time dependent study. Our problem was a steady state- steady flow so we selected a stationary study.

Next we selected material and defined the boundary conditions. For this simulation, air was chosen as the material to be used for modeling of the fluid flow inside the DBD reactor due to the high dilution of exhaust gases with air in experiments. The last step in setting up the physical parameters of the model was to set up the boundary conditions. The boundary conditions that were relevant in this model were fluid properties, inlet and outlet and wall boundaries and were applied to each of the four edges in the model. For the inlet boundary, laminar inflow was used with a 1 m long entrance length. This meant the reactor was extended theoretically for 1m to ensure the flow entering the reactor was fully developed. The outlet boundary was set with a gauge pressure of 0 to simulate open air conditions. The wall boundaries were set up under no slip conditions to simulate physical walls.

Finally the fluid properties were set to being air at 300K and a pressure of 1atm. COMSOL offers the user an automated meshing function which sets up a mesh based on the physics chosen in the model. In this case, automated meshing function produced a fairly fine mesh made up triangular elements and had a greater density near the wall boundaries. The basic configuration of the reactor and the axisymmetric model which was used by COMSOL is shown in Fig. 1.

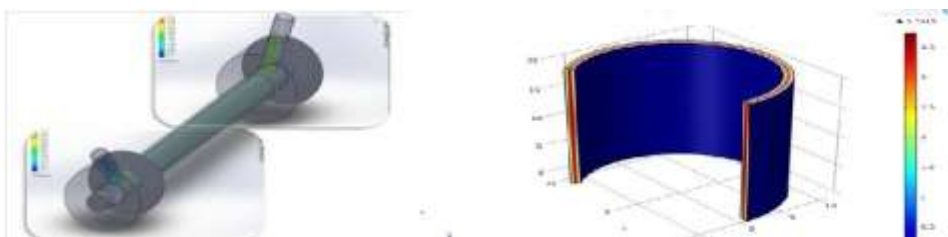


Figure 1. Reactor configuration and axisymmetric model used in COMSOL

Residence time distribution

RTD is a crucial parameter when treating engine exhaust emissions with a DBD reactor. If the mean residence time increases in a DBD reactor, there will be a corresponding increase in reaction time and consequently, the pollutant removal efficiency can increase.

The RTD in ideal plug flow reactors is defined as the ratio of the volume to the volumetric flow rate ($\frac{Volume}{Volumetric\ flow\ rate}$). This can be used as an estimation for real RTD in all other reactors. The RTD is determined by injecting an inert tracer into the reactor at some time $t=0$ and then measuring the tracer concentration, C , in the effluent stream as a function of time. There are two methods of injecting tracer into a reactor: pulse input and step input. For the scope of this project, pulse input was explored and discussed in the following section. In a pulse input, an amount of tracer, N_0 , is suddenly injected in one shot into the feed stream entering the reactor as short time as possible. The outlet concentration is then measured as a function of time. The amount of tracer material, ΔN , leaving the reactor between time t and $t+\Delta t$ is then:

$$\Delta N = C(t)V\Delta t \quad (1)$$

where V is the volumetric flow rate. When the total amount of material that was injected into the reactor, N_0 , is divided, the following equation can be obtained.

$$\frac{\Delta N}{N_0} = \frac{C(t)V}{N_0} \Delta t \quad (2)$$

which represents the fraction of material that has a residence time in the reactor between time t and $t+\Delta t$. For pulse injection it can be defined as follow:

$$E(t) = \frac{C(t)V}{N_0} \quad (3)$$

The quantity $E(t)$ is called the residence-time distribution function. It is the function that describes in a quantitative manner how much time different fluid elements have spent in the reactor. If N_0 is not known directly, it can be obtained from the outlet concentration measurements by summing up all the amounts of materials, ΔN , between time equal to zero and infinity. Writing Equation 1 in differential form gives

$$dN = VC(t)dt \quad (4)$$

And then by integrating gives

$$N_0 = \int_0^{\infty} VC(t)dt \quad (5)$$

The volumetric flow rate v is usually constant, hence $E(t)$ can be defined as:

$$E(t) = \frac{C(t)}{\int_0^{\infty} C(t)dt} \quad (6)$$

As is the case with other variables described by distribution functions, the mean value of the variable is equal to the first moment of the RTD function, $E(t)$. Thus the mean residence time (τ) is:

$$\tau = \frac{\int_0^{\infty} tE(t)dt}{\int_0^{\infty} E(t)dt} = \int_0^{\infty} tE(t)dt \quad (7)$$

In this paper, COMSOL Multiphysics software solved the concentration equation with the convection and diffusion terms to model the tracer and calculate the residence time based on that (COMSOL.Model.Gallery 2008). The concentration equation is shown in Eq.8:

$$\frac{\partial c}{\partial t} + \nabla \cdot (-D\nabla c + cu) = 0 \quad (8)$$

Where c denotes the concentration ($\frac{\text{kg}}{\text{m}^3}$), D is diffusion coefficient ($\frac{\text{m}^2}{\text{s}}$), and u refers to the velocity ($\frac{\text{m}}{\text{s}}$). The velocity is given by steady state solution of Navier-Stokes equations which has been described in previous section. To simulate the tracer, a time-dependent boundary condition at the inlet can be employed:

$$c = c_0 e^{-(t-3)^2} \quad (9)$$

Where $c_0 = 1(\frac{\text{mol}}{\text{m}^3})$. Based on this equation, the mean of the inlet pulse function is 3 s. By calculating the integral of the concentration at the reactor outlet and using Eq. 7, the mean of the outlet pulse function can be determined. The difference in time, that is the time at which the pulse appears at the outlet minus the time at which the pulse enters the reactor, will be the mean residence time of the reactor. The boundary condition at the outlet is a convective-flux condition stating that all mass transport over the boundary occurs through convection. All other boundaries are insulated. A 2D axisymmetric model of a DBD reactor is developed for the residence time investigation as described above.

Results and discussions

Our experiments are based on a reactor with 1mm, 2mm and 3mm gaps. These gaps are appropriate for producing plasma. If we increase the gaps too much, it is not easy to produce plasma and the amount of required electrical energy increases substantially. In all models, inlet velocity with the value of 2.5 m/s is the base case because our initial experimental tests for emission reduction have been done for this velocity. First we fixed the flow rate (i.e. inlet velocity) and studied the effect of gap increase in our model. Inlet velocity kept constant at 2.5 m/s and gaps varied from 1mm to 3mm. the effect of different gap sizes on RTD is shown in three graphs in Fig. 2.

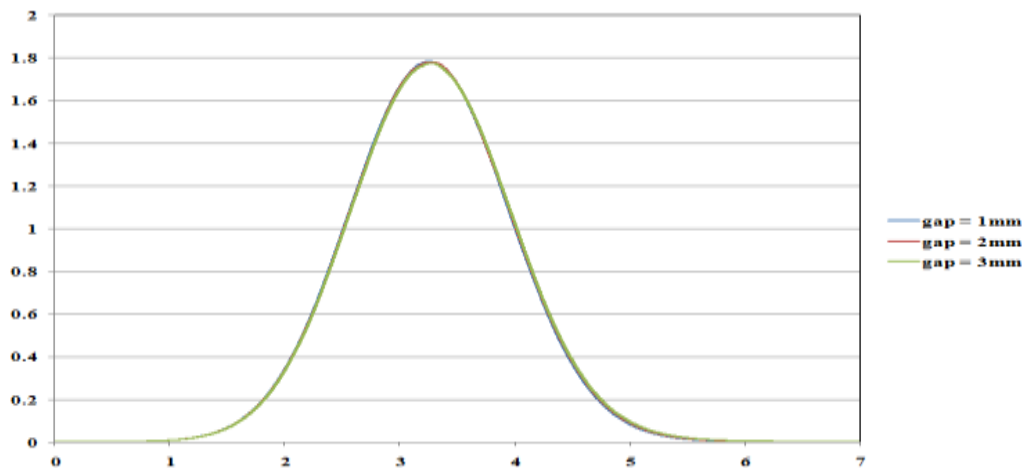


Figure 2. Comparison of RTD for different gap sizes in reactor outlet

These three graphs are almost similar and look to be coincidence on each other in Fig. 2. The time at which the pulse appears at the reactor outlet can be calculated from this figure. It will be 3.104353s, 3.112467s and 3.117679s for 1mm, 2mm and 3mm respectively. These times minus the time at which the pulse enters the reactor (3s) will be the mean residence time. Therefore the

average RTD for 1mm, 2mm and 3mm gaps are 0.104353, 0.112467 and 0.117679 respectively. As we expected by increasing the gap size, average RTD increases.

Then we are interested in studying the effects of different flow rate on RTD. Five different velocities have been considered (0.025 m/s, 0.25 m/s, 2.5 m/s, 5 m/s and 25 m/s). The flow is considered to be laminar in all velocity except for $u=25\text{m/s}$ which has been modeled as turbulent flow.

Table 1. Effect of different velocity on residence time distribution

Velocity (m/s)	τ (Modeling results) (s)	Percentage of increase in τ compare to base case (%)	τ (Ideal plug flow) (s)
0.025	8.922487	8450	7.916716146
0.25	0.800073	666.7	0.791671615
2.5	0.104353	(Base case)	0.079167161
5	0.040993	-60.72	0.039583581
10	0.008515	-91.84	0.007916716

By increasing velocity from $u=2.5\text{ m/s}$ to $u=5\text{ m/s}$ and $u=25\text{ m/s}$, the mean residence time gradually decreases. But there is a significant increase in residence time when velocity decreases to 0.25 m/s and .025 m/s. For a better understanding of velocity effect on RTD, average residence time for these different velocities (flow rates) has been calculated and summarized in Table 1 above.

One of the conventional methods for improving RTD is adding some inside the reactor. To investigate the effect of the number of roughness elements on RTD, the geometry of the gap has been revised and shown in Fig.3 below. From this figure, we can see that “a” represents the height of the baffle, “b” represents the width of the baffle and “t” represents the distance between two baffles. These are the variables that we need to test for the flow simulations and study the effect of them on residence time.

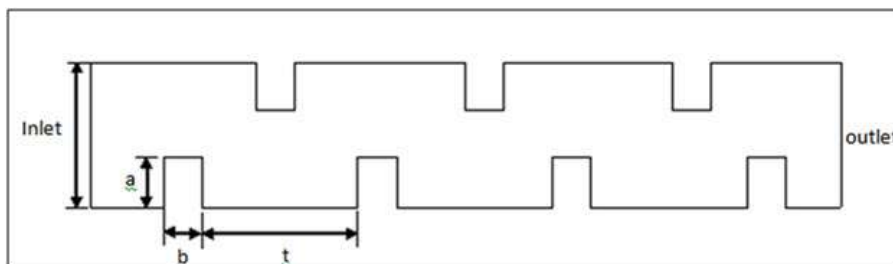


Figure 3 .Revised Geometry of gap for study the effect of the roughness elements on RTD

We considered the gap of 3mm for roughness elements RTD simulation. This gap was bigger and there was the potential to add some roughness elements in real applications. First we studied the effect of number of the roughness elements on RTD. In this step of simulation, the gap was 3mm and velocity was 2.5 m/s, roughness element's height and width were 1.5mm and 1mm respectively. We changed the distance between roughness elements in our simulation to study the residence time

distribution. By adding roughness elements, the cross section of the flow passage decreased and velocity increased. This reduced the residence time of flow inside the reactor. On the other hand, RTD by adding roughness elements increased due to the enlargement of the flow path area inside the reactor. These effects contrasted with each other. Therefore there should be an optimum in number of roughness elements.

Table 2. Effect of the roughness elements spacing on RTD ($u=2.5\text{m/s}$, $\text{gap}=3\text{mm}$)

Variant	a (mm)	b (mm)	t (mm)	No. of baffles on outer wall	No. of baffles on inner wall	Mean residence time(s)	Pressure drop (Pa)
Without roughness	n/a	n/a	n/a	n/a	n/a	0.117679	12.432
Case 1	1.5	1	19.5	10	9	0.097282	194.91
Case 2	1.5	1	29.5	7	6	0.129283	104.64
Case 3	1.5	1	39.5	5	5	0.147425	82.435
Case 4	1.5	1	49.5	4	4	0.162413	73.512
Case 5	1.5	1	59.5	4	3	0.151238	59.831

As we can see in Table 2 above, the number of roughness elements was too many in case 1 and the average RTD is lower than the base case without any roughness. When the number of roughness elements reduced to 4 in case 4, we found the optimum of RTD in our simulation. Further reduction in number of roughness elements decreased the average RTD. The maximum of RTD was around 0.16 s in case 4 and it was around 38% bigger than average RTD for the base case. This increase in RTD can have a considerable effect on plasma emission reduction without producing any significant pressure drop (Okubo, Kuroki et al. 2003).

Table 3. Effect of baffle spacing on RTD ($u=2.5\text{m/s}$, $\text{gap}=3\text{mm}$)

Variant	a (mm)	b (mm)	t (mm)	No. of baffles on outer wall	No. of baffles on inner wall	Mean residence time (s)	Pressure drop (Pa)
Without roughness	n/a	n/a	n/a	n/a	n/a	0.117679	12.432
Case 1	1	1	19.5	10	9	0.123512	47.085
Case 2	1	1	29.5	7	6	0.150882	36.564
Case 3	1	1	39.5	5	5	0.216638	32.202
Case 4	1	1	49.5	4	4	0.285038	29.119
Case 5	1	1	69.5	3	3	0.336134	25.92
Case 6	1	1	79.5	3	2	0.340349	23.766
Case 7	1	1	89.5	3	2	0.357117	26.75
Case 8	1	1	99.5	2	2	0.371819	20.925
Case 9	1	1	109.5	2	2	0.38668	21.372
Case 10	1	1	119.5	2	2	0.396263	22.689
Case 11	1	1	129.5	2	1	0.404621	19.145

Case 12	1	1	139.5	2	1	0.414272	19.227
Case 13	1	1	149.5	2	1	0.422341	19.246
Case 14	1	1	159.5	2	1	0.450031	19.871
Case 15	1	1	169.5	2	1	0.43875	20.615
Case 16	1	1	179.5	2	1	0.441192	23.139
Case 17	1	1	189.5	1	1	0.447328	16.774

We decreased the roughness elements height (a) to 1mm and studied the effect of roughness elements numbers on RTD as shown in Table 3 above. The trend was the same as before. When the number of roughness elements was too many (case 1), the roughness elements are not so effective. When the number of roughness elements decreased to less than three on one side of the reactor, we can consider different configurations with the same number of roughness elements and different roughness elements spacing (t). One of the interesting results was obtained in case 17. By using just 2 baffles, one in reactor inlet and another one in the reactor outlet, we could get the residence time of 0.447328 which was four times bigger than the case without any roughness. This trend has been observed for 1mm gap and 1/3 mm baffle height simulation as well. Due to the length limitation of this paper, those results are not represented in this paper.

Fig. 4 represents the velocity contour inside the reactor for just one of the roughness elements with 1.5 mm height and 3mm gap. The contour in blue indicates the minimum velocity while red indicates the maximum velocity. When flow passed a roughness element, velocity increased due to the decrease of the cross section. The maximum velocity was 7.8397 m/s based on this simulation by COMSOL.

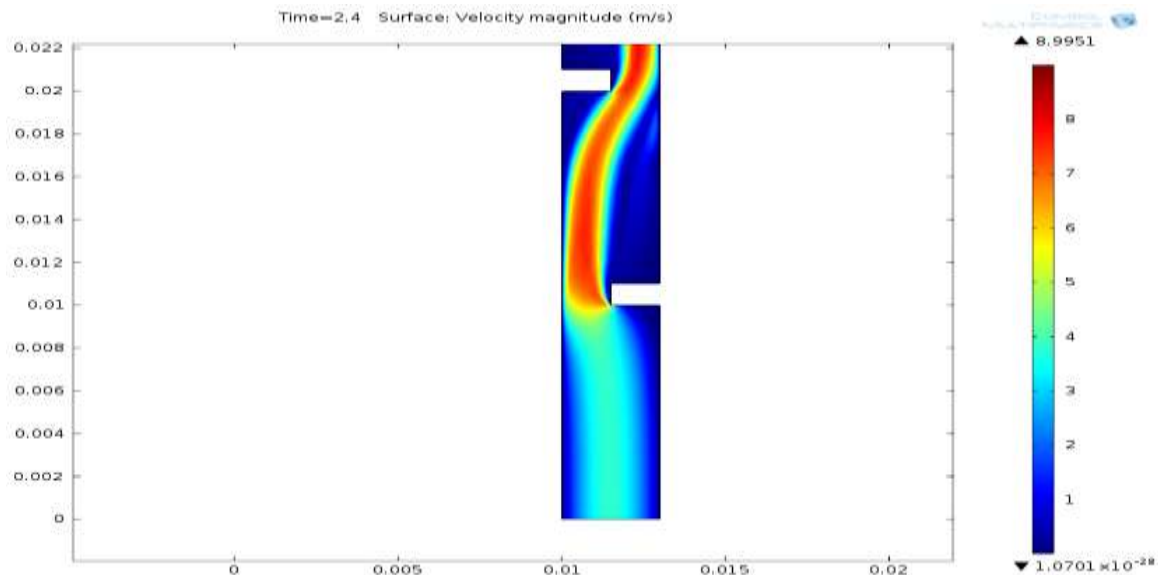


Figure 4. Velocity contour of flow inside the reactor

Grid dependency study

CFD simulation of mean residence time has been evaluated for different grid numbers and the results are shown in Fig. 5. As we can see, there is no significant grid dependency when the number of mesh elements is greater than 36000.

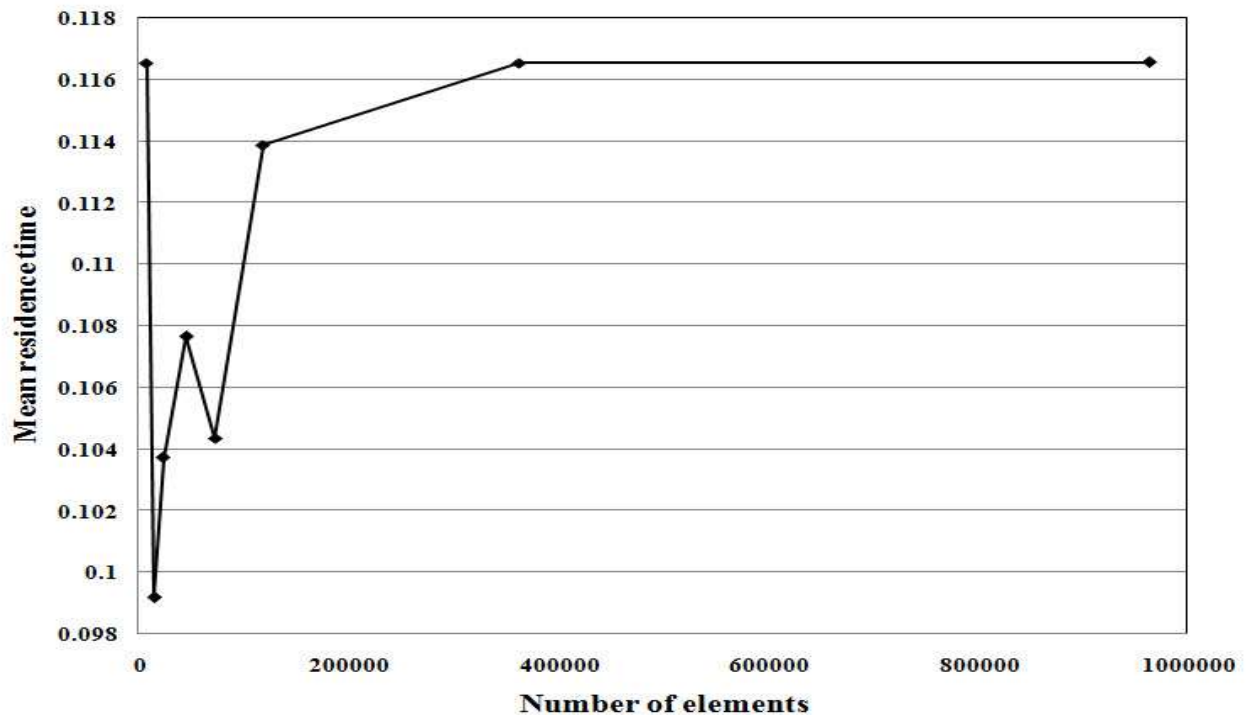


Figure 5. Grid dependency analysis of RTD

Conclusion

Residence time distribution of a conventional DBD reactor has been modelled and the effect of different roughness elements has been studied. It was shown that adding appropriate roughness elements can increase the residence significantly without any considerable pressure drop. These results will form the basis for a COMSOL plasma and CFD module investigation in future modeling and experiments.

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