Research on the Distribution of Vaporized Hydrogen Peroxide Based on UDF and Experimental verification

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Abstract. In order to obtain the concentration distribution rule of vaporized hydrogen peroxide during sterilization, UDF (user-defined function) is used to describe the functional relationship of the vaporized hydrogen peroxide mass fraction at the equipment inlet, Using the software AnsysFluent, the flow field in the space is mathematically simulated to obtain the mixture flow field. The comparative analysis with the data measured from field experimental verified the correctness of the simulation results. Studies have shown that vortices appear in the flow field near top of the room and wall surface; the concentration of vaporized hydrogen peroxide increases with the height of the space; Elevated concentration of vaporized hydrogen is found at top of the sterilization chamber, and its content is higher than the middle and bottom of the sterilization chamber. Using UDF to describe concentration of the hydrogen peroxide at the entrance of the equipment is in line with the actual working conditions and can improve the simulation accuracy.

1. Introduction
Vaporized hydrogen peroxide has a strong sterilization ability[1]. After sterilization it is decomposed into water and oxygen, which is more efficient and environmentally friendly than traditional sterilization technology[2]. Vaporized hydrogen peroxide has been widely used in biopharmaceutical medical and healthcare fields[3] [4] [5]. At present, there are a lot of research on vaporizing hydrogen peroxide. Ren Zhe [6] and others have found that bacteria on stainless steel and plastic are more difficult to kill than bacteria on glass, indicating that the sterilization effect will be affected by the material. Qi Jiancheng [7] and others invented a flash steaming method, which can make vaporized hydrogen peroxide diffuse evenly. Ye Kaixue [8] used high-frequency resonant vibrating plates to separate gaseous hydrogen peroxide at only about 40 degrees Celsius. Zeng Shiqing [9] used a high-frequency sound wave generator to generate micron-level floatable hydrogen peroxide particles.

Although there are many studies focus on vaporized hydrogen peroxide equipment, few studies focus on the flow field distribution in the sterilization space, and few studies are verified with field experiment. Therefore, exploring the distribution of concentration flow field and the methods to measure it are of great significance for studying the uniformity of diffusion. In this paper, the numerical simulation of the concentration distribution of vaporized hydrogen peroxide in the sterilized space is carried out, and combined with the field experimental data, the distribution of hydrogen peroxide...
concentration in the sterilized space is obtained. It can provide a reference for the optimal design of the equipment, thereby further improve the sterilization effect and efficiency.

2. Mathematical Model

2.1. Continuity equation

Based on Species Transportation model simulated by Ansys Fluent, the system inlet is composed of high temperature air, vaporized hydrogen peroxide and water vapor. A standard k-ε model is used. The mixture at the inlet diffuses into the air at room temperature in the sterilized space, and the free path of the molecule is at the nanometer level, satisfying the condition of no slip. Gases are considered incompressible fluids. The detailed k-ε turbulence continuity equation is as follows [10]:

\[
\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho u_i) = 0, \quad i = 1, 2, 3
\]

In the formula: \(x_i\)—displacement, \(\rho\)—density, \(t\)—time; \(u_i\)—component velocity. Using Renault pressure and Renault mean speed to get [11]

\[
-\rho u_i = \mu \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right) - \frac{2}{3} \left( \frac{\partial k}{\partial x_i} + \frac{\partial (\mu \frac{\partial u_i}{\partial x_j})}{\partial x_j} \right) \delta_{i,j}, \quad i, j = 1, 2, 3
\]

In the formula: \(c_{\mu}\)—constant, \(k\)—turbulent kinetic energy, \(u_i\)—component velocity; \(\delta_{i,j}\)—Kronecker operator. The \(k-\varepsilon\) turbulence continuity equation

\[
\frac{\partial k}{\partial t} + \nabla \cdot \left( \rho \frac{\partial k}{\partial x_j} \right) = C_1 \frac{\varepsilon}{k} G_k - C_2 \frac{k}{\varepsilon} Y_m + \frac{1}{\sigma_k} \frac{\partial}{\partial x_j} \left( \sigma_k \frac{\partial k}{\partial x_j} \right) + C_2 \frac{G_k^2}{k} - C_3 \frac{k^2}{k}
\]

In the formula: \(\mu\)—fluid viscosity, \(\varepsilon\)—dissipation rate; \(\sigma_k, \sigma_z\)—Prandt number, \(G_k\), and \(G_b\)—turbulent flow energy; \(Y_M\)—fluctuation caused by turbulent diffusion; \(C_{1z}, C_{2z}, C_{3z}\) are constants.

2.2. Energy conservation equation

Since components in the flowing medium sprayed by the equipment do not have chemical reaction with each other, the species transport model and the conservation equation of matter are used for calculation. The conservation equation of matter:

\[
\frac{\partial}{\partial t} \left( \rho \omega \right) + \nabla \cdot \left( \rho u_i \omega_i \right) = \nabla j_i + R_i + S_i, \quad i = 1, 2, 3
\]

In the formula: \(J_i\)—Diffusion flux of matter; \(R_i\) and \(S_i\) are the net production rate of the chemical reaction and the additional production rate caused by the source phase [12] [13]. Since no chemical reaction is considered, \(R_i\) and \(S_i\) are 0. The System of equations composed of the above equations can be obtained by using Fluent to calculate the domain extent after meshing to obtain the flow field such as the concentration field of the control domain.
2.3. Boundary conditions
1. The walls of the room and equipment to be sterilized are non-slip walls, with a symmetrical surface; 2. Application of standard k-ε model [14][15]; 3. Satisfying open energy equation; 4. Establishing a transport model for non-reactive components; 5. Adding gravity acceleration; 6. Defining the physical properties such as the density of each component and treat it as a constant [16] and then using UDF to compile the inlet component concentration; 7. Using transient simulation of the model. 8. Using SIMPLE algorithm. 9. Using velocity inlet [17]. At this time, the inlet velocity of vaporized hydrogen peroxide is 4.5 m/s, and the inlet adopts Species Transport. 10. The pressure outlet is used for the system outlet.

3. The goodness of fit RNew
In order to study the accuracy of UDF description of the actual working conditions of the nozzle and the accuracy of numerical simulation, the fitting goodness of fit RNew is used for analysis. which can reflect the fitting degree of the non-linear curve. The equation is:

\[
R_{New} = 1 - \sqrt{\frac{Q}{\sum (y-y^*)^2}}
\]

(6)

\[
Q = \sum (y-y^*)^2
\]

(7)
y—actual measured value, \(y^*\)—predicted value. The closer RNew is to 1, the better the curve fit.

4. Geometric model
4.1. Project Overview
The main tasks of the vaporized hydrogen peroxide sterilization equipment are: first mix the air and the hydrogen peroxide solution, atomize by the siphon effect of the Venturi tube, and then use the evaporating dish to heat and vaporize the hydrogen peroxide, finally using the equipment to spray the vaporized hydrogen peroxide into the space to sterilization. However, the equipment nozzle will re-accommodate part of the hydrogen peroxide in the sterilized space, making its working conditions complicated. The sterilization space is composed of a simple brick house building, 3.5 m high, 4 m long, and 5 m wide. The vaporized hydrogen peroxide sterilization equipment is placed in the center of the space to provide a continuous stream of vaporized hydrogen peroxide for sterilization of the space.

4.2. Geometric model establishment and meshing
According to the relative position and size of the sterilization space and the sterilization equipment, Solidworks is used to establish an approximate 1:1 geometric model, the sterilization equipment is appropriately simplified, and the nozzles and other structures of the sterilization equipment are regarded as relatively regular. The shape and internal structure of the equipment are not considered further, mainly simulating the movement law and spatial distribution of hydrogen peroxide in the sterilization space. Since the geometric model has symmetry, it is sufficient to build half of the complete model. Use SolidWorks to create a geometric model and import it into Ansys, and divide the calculation grid, as shown in Figure 1.
5. Geometric model

5.1. Using UDF to simulate the volume fraction function of hydrogen peroxide in the nozzle

The atomization and vaporization of the hydrogen peroxide solution in the equipment are carried out inside the equipment, but as the equipment continues to work, the equipment will absorb some hydrogen peroxide sterilized rooms and continue to spray. so that its concentration will continue to increase as the sterilization process. It’s difficult for the conventional method to accurately describe the concentration relationship of the inlet hydrogen peroxide. Therefore, by first assuming that the system entrance hydrogen peroxide is a constant, using fluent to perform sterilization simulation on the sterilized space, the function of the system outlet vaporized hydrogen peroxide mass fraction with respect to time is obtained, and then using Matlab to obtain its mathematical relationship, As shown in Figure 2. Finally, udf is used to simulate the functional relationship of the mass fraction of vaporized hydrogen peroxide at the inlet and outlet.

\[
 f(t) = a_0 + a_1 \cos(wt) + b_1 \sin(wt) + a_2 \cos(2wt) + b_2 \sin(2wt) 
\]

\(a_0=0.000354, a_1=-0.0004555, b_1=0.001098, a_2=0.0001015, b_2=-0.0001186, w=0.9738.\) t— time, and f(t) is the mass fraction of vaporized hydrogen peroxide in compressed air in this state. Through further
calculation, the functional relationship of the mass fraction of vaporized hydrogen peroxide at the inlet can be obtained.

5.2. Steam flow field and vaporized hydrogen peroxide concentration distribution
During the sterilization process of the equipment, the vaporized hydrogen peroxide enters the sterilized space at a certain wind speed. The wind speed is 4.5 m/s. The air flow in the sterilized space is numerically simulated to obtain the vector diagram and streamline of the steam flow inside the space. It can be seen from the figure 3: A large vortex appears on the top of the sterilization chamber, and a small part of the mixture is absorbed by the equipment.

![Figure 3. Streamline diagram of the sterilized room](image)

6. Comparison of measured data and simulation results

6.1. Test method and field measurement
According to the sampling point arrangement method in the related literature, it is shown in figure 5.

![Figure 4. Arrangement of spatial measuring points and Field test](image)
Measurement points 1 and 2 and point 3 are arranged in the room to be sterilized. Each measurement point is equipped with a vaporized hydrogen peroxide concentration sensor. Before the test, we use a thermostatic equipment to keep the room temperature at a constant 20 degree Celsius, and apply desiccant to the sterilized room, preventing the temperature and humidity from interfering with the diffusion of vaporized hydrogen peroxide in the room. Turning on the heating device first to ensure that the atomized hydrogen peroxide can be vaporized directly. Adjusting the flow rate of the peristaltic pump and air compression pump to ensure that the flow rate of the hydrogen peroxide solution is 10ml/min and the air compression amount is 7.5L/min. Then the actual measurement of the concentration of vaporized peroxide, as shown in Figure5, recording data every 2 minutes. After the test data are collected at each data collection point, the remnant in the room are immediately cleared. Each test is repeated 3 times at each data collection point and average of measurements are used in analysis. Compare the measured value of the vaporized hydrogen peroxide concentration with time to the value of the concentration change at the corresponding point in the space using the UDF to describe the inlet vaporized hydrogen peroxide content and the inlet hydrogen peroxide as constants in the simulation, as shown in Table 1.

**Table 1. Variation of vaporized hydrogen peroxide concentration with time at each measurement point**

| Measure point | Concentration (ppm) | Time(min) |
|---------------|---------------------|-----------|
|               | 0 2 4 6 8 10 12 14 16 18 20 |           |
| Field | | | | | | | | | | |
| measurement | 0 0 0 1.3 9.3 14.4 22.4 30.4 45.0 55.2 65.6 | |
| Use UDF | 0 0.3 2.8 7.1 10.4 18.3 24.5 31.7 39.7 48.6 56.3 | |
| inlet is constant | 0 0.3 2.6 6.5 11.3 16.6 21.9 27.3 32.6 37.9 46.0 | |
| Field | 0 0 0 6.2 13.3 16.4 24.7 34.7 50.4 56.7 69.7 | |
| measurement | 0 1.9 5 10.1 16.9 23.2 28.7 34.5 41.2 53.8 63.8 | |
| Use UDF | 0 1.5 4.2 9.3 15.8 19.5 25.0 30.4 35.8 45.2 56.5 | |
| inlet is constant | 0 0 0 57.3 99.9 145.4 194.6 246.4 292.1 341.5 370.3 390.8 | |
| Field | 0 0 57.3 99.9 145.4 194.6 246.4 292.1 341.5 370.3 390.8 | |
| measurement | 0 47.7 91.1 130.7 170.5 205.6 242.8 277.8 310.8 341.9 361.4 | |
| Use UDF | 0 39.7 82.0 112.9 151.5 177.2 200.4 226.4 262.5 287.3 300.8 | |

The concentration of vaporized hydrogen peroxide in the room to be sterilized increases with the injection time. The highest concentration is at measurement point No. 3, locating at the top of the room, and the lowest concentration is measured at measurement point 1, locating at bottom of the room. It is located at the bottom of the sterilized room. The distribution of vaporized hydrogen peroxide in the whole room is not uniform, mainly concentrated on the top of the room.
6.2. Comparison of simulation results with actual measurement results
According to the comparison between the measured hydrogen peroxide concentration and the numerical simulation results of the vaporized hydrogen peroxide mass concentration in the room to be sterilized, drawing the curve of the vaporized hydrogen peroxide concentration with time for each operating condition and measurement point. The comparison between the two is shown in Figure 6-8. The concentration of vaporized hydrogen peroxide at the air inlet increases with the increase of the injection time. The inlet concentration is used as a constant in the measurement points 1, 2, and 3. The numerical curve of the hydrogen peroxide mass concentration obtained from the simulation and the actual measured value curve, through the fit index RNew is 79.93%, 82.89%, 77.61% respectively, using UDF to describe the simulation curve of the inlet concentration, the fitting index RNew is 92.87%, 94.07%, 93.06%, indicating the use of UDF simulation curve to modify the change of the import concentration, it reflects the actual working conditions very well. After 1 iteration, the error is reduced, so this correction method is correct. If need to improve the accuracy, increase the number of stacking times.

Figure 5. Curves of vaporized hydrogen peroxide concentration (ppm) with time (t/s) of three different methods in measuring point one

Figure 6. Curves of vaporized hydrogen peroxide concentration (ppm) with time (t/s) of three different methods in measuring point two
At the beginning of the actual sterilization, due to the presence of a small amount of bacteria and microorganisms in the space, the hydrogen peroxide sprayed from the equipment will combine with it and decompose, and the equipment will also cause a small amount of hydrogen peroxide to decompose during the vaporization process. This makes the hydrogen peroxide vaporization concentration in the actual measurement results in the early stage of sterilization lower than the simulation results. And in the actual sterilization process of the equipment itself, the compressed air contains the sprayed hydrogen peroxide and the solution is mixed and atomized, so that the content of vaporized hydrogen peroxide sprayed from the nozzle is getting higher and higher. When UDF describes the injection conditions at the inlet, the vaporized hydrogen peroxide and hydrogen peroxide solution re-inhaled by the entire system are calculated by superposition, which makes the injection conditions simulated by UDF slightly different from the actual ones. The concentration of vaporized hydrogen peroxide in the measured results at the end of sterilization is higher than the simulated results. At the same time, the data obtained by simulating the inlet peroxidation as a constant will be lower than the data using UDF to describe the inlet injection conditions.

7. Conclusion

1. Through the simulation and actual measurement of the 20-minute sterilization process, the distribution law of hydrogen peroxide concentration was obtained: the steam flow inside the sterilized sterilization chamber is disordered, and there are vortex areas between the walls. The top concentration is within 370ppm-450ppm, while the bottom concentration is within 60ppm-70ppm, the concentration of vaporized hydrogen peroxide is not evenly distributed in the room.

2. Use UDF to modify the inlet hydrogen peroxide concentration to obtain a vaporized hydrogen peroxide concentration change curve. The fit index between the curve and the measured curve R_new is more than 93.06%. It can be seen that using UDF to modify the inlet concentration can effectively improve the simulation accuracy.

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References

[1] Otter, J. A., Yezli, S., & French, G. L. (2012). Impact of the suspending medium on susceptibility of meticillin-resistant Staphylococcus aureus to hydrogen peroxide vapour decontamination. Journal of Hospital Infection, 82(3), 213-215. doi:10.1016/j.jhin.2012.08.006

[2] Frey, H. E., & Pollard, E. C. (1966). Ionizing Radiation and Bacteria: Nature of the Effect of Irradiated Medium. Radiation Research, 28(3), 668. doi:10.2307/3571994
[3] Yao Dongling. Current Situation and Prospect of Hydrogen Peroxide Production in China [J]. Inorganic Salt Industry, 2013, 45(09): 1-4+23.

[4] He Jiangfeng. Production method and application of hydrogen peroxide [J]. Hebei Chemical Industry, 2009, 32(05): 9-12.

[5] Fu Chengfeng. Comparing the effect of using hydrogen peroxide sol spray and peroxycetic acid aerosol spray to disinfect the ward air [J]. Contemporary Medicine Collection, 2016, 14(03): 166-167.

[6] Ren Zhe, Wei Qiuhua, Rao Lin et al. Research on the killing effect of hydrogen peroxide vapor sterilizer on the surface of different materials [J]. Chinese Journal of Disinfection, 2015(03): 214-216.

[7] Qi Jiancheng, Chen Xu, Wu Jinhui, etc. A hydrogen peroxide steam sterilization device [P]. Chinese Patent, CN 103182093 U, 2013-7-3.

[8] Ye Kaixue. A hydrogen peroxide low temperature vaporization sterilization device and sterilization method [P]. Chinese Patent, CN 105056265 A, 2015-11-18.

[9] Zeng Shiqing. A hydrogen peroxide vaporization method and vaporization device [P]. Chinese patent, CN 105727330 A, 2016-7-6.

[10] Liu Bo, Gao Ge. Study on the characteristics of quasi-order structure of turbulent boundary layer using GAO-YONG equations [J]. Journal of Aerodynamics, 2003(03): 313-321.

[11] Sun Liqiang, Hu Yue, Wang Di, Song Jianfei, Wei Yaodong. Comparative analysis of dynamic characteristics of RSM and LES simulated cyclone separator flow field [J]. Chemical Reaction Engineering and Technology, 2018, 34(04): 289-296.

[12] Konishi T, Akagi S, Kikugawa H, et al. Flow Visualization of Waste-Heat Boiler Using 1/20 Scale Model and Numerical Simulation [M]// Progress in Scale Modeling. Springer Netherlands, 2008.

[13] Zhang Jingyu. Research on the suppression of infrared radiation of thermal jet by spherical discrete particles [D]. Nanjing University of Aeronautics and Astronautics, 2007.

[14] Wang Handing, Cheng Zhengzai, Tang Ming, Huang Zhengliang, Yang Yao, Yang Yongrong. Unsteady state analysis of polypropylene loop reactor based on CFD simulation [J]. Chemical Reaction Engineering and Technology, 2018, 34(05): 424-430.

[15] Tan Shengkui, Wang Rui, An Ruidong, Li Jia. Research and Application of Mathematical Model of Muddy Water Density Flow Based on Component Transport Model and RNG k-ε Model [J]. Journal of Sichuan University (Engineering Science Edition)), 2011, 43(S1): 48-53.

[16] M Chen, J Aleixo, S williams, el al, CFD modelling of 3-Way Catalytic Converters with Detailed Catalytic surface Reaction echanism

[17] Lan Shaolian, Zhu Dongsheng, Zhong Jiasong, Zhao Qiang. Numerical simulation of corrugated plate heat exchanger [J]. Chemical Engineering, 2010, 38(08): 18-21.