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STUDY OF GREEN HYDROGEN-AIR MIXTURE FORMATION IN A CLOSED CHAMBER WITH A GIVEN COMPOSITION

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The subject of this study is the formation of a hydrogen-air fuel mixture with a given composition and the required degree of homogeneity in a closed chamber while taking into consideration conditions for generating hydrogen by an electrolytic method. This study includes an industrial technology fueled by combustible gases to process thermoplastic parts. The motivation behind this research is the transition to carbon-free processing technology, which would not only reduce emissions and is in line with the concept of a climate-neutral industry but also raises the level of safety with regard to these technologies to a high level. One of the main conditions of high-quality thermal processing fueled by combustible gases is still the precise quality control of the composition of the mixture and its operating pressure. Based on the specific requirements of the researched technology, a mathematical model for generating fuel mixtures was developed, which determines the characteristics of a multi-component gas mixture while the working chamber is filling and during the holding period after the gas supply has stopped. The developed mathematical model is supplemented with criteria for evaluating the quality of the fuel mixture. A numerical study using computational fluid dynamics (CFD) was conducted to assess the specified criteria required to achieve a quality fuel mixture.

Keywords: green hydrogen, fuel mixture, evaluation criteria of fuel mixture, numerical simulation, mixture generation

1. Introduction

The research area is inspired by the European Green Deal under the New Industrial Strategy and Climate and Energy Policy Framework, as well as the SRRI Roadmap (2021), aiming to improve energy efficiency and reduce greenhouse gas emissions from industrial processes. This research covers industrial technologies fueled by combustible gases to process thermoplastic parts and possibly replace energy sources based on fossil fuels (coal and methane) with hydrogen produced using electricity. Such a transition to carbon-free processing technologies would not only reduce emissions and help to facilitate a climate-neutral industry but ensure the level of production safety of these technologies is high.

The goal behind the researched technology, based on the Thermal Energy Method (TEM), is the final processing of various materials, including plastics [1]. At present, engineering plastics and high-performance plastics (EP and HPP) - due to their low weight, high strength and corrosion resistance - are widely used in the automotive and aerospace industries, in medicine, in numerous electronic gadgets and devices as well as in power electronics. The production of plastic parts requires thermal polishing, curing the pores of thermoplastic structures and removing surface defects.

Currently, TEM is applied to hydrogen-air mixtures with an excess of hydrogen for removing burrs, fogging and the partial surface polishing of parts made of some thermoplastic materials, e.g. acrylic, polyurethane, polyethylene, etc. [2]. Stoichiometric mixtures or mixtures containing an excess of fuel are best suited for the TEM processing of thermoplastic parts. According to [3], the ratio of oxygen to hydrogen in the fuel mixture should be 1:3 (with a stoichiometric ratio of 1:2). However, these statements relate to oxygen-hydrogen mixtures rather than the air-hydrogen mixtures under study, neglect the effect of the composition of the mixture on the energy capacity as well as do not take into account the presence of the treated object in the combustion chamber nor its properties. As a result, they must be refined to ensure the predicted quality of treatment. In light of this, this research investigates the formation of a green hydrogen-air mixture in a closed chamber in order to establish the best way to form a fuel mixture with a given composition and the required degree of homogeneity while taking into account the conditions

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necessary to generate green hydrogen. An additional task in this regard is to select an effective way of obtaining green hydrogen. The issue of generating green hydrogen is quite well studied. In works [4] and [5], hydrogen production technologies, storage technologies as well as the end-use applications of hydrogen based on the source of the input energy, operating conditions, conversion efficiency, energy density and unit investment costs were comprehensively reviewed, moreover, the advantages and disadvantages, technological readiness of hydrogen production technologies in addition to storage methods highlighted. The analysis of the indicated data proves the possibility and rationality of obtaining green hydrogen for the technology studied in an electrolytic way by directly using an electrolyzer with a known level of productivity [6].

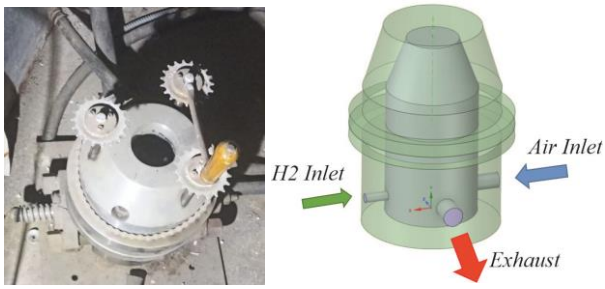
The main factors affecting the quality of plastics processed by TEM include the composition of the mixture, initial pressure, ignition method, combustion modes and time during which parts are in contact with combustion products [7]. Due to the large number of parameters that affect the development of the specified patterns, it is advisable to set them consistently and in a connected manner in an analytical way.

Therefore, within the framework of this work, a mathematical model of the generation of a hydrogen-air mixture is developed to determine the characteristics of a multi-component gas mixture. Moreover, numerical studies of the filling process are carried out by taking into account the conditions required to accurately determine the mixture of the composition and its degree of homogeneity.

2. Problem statement

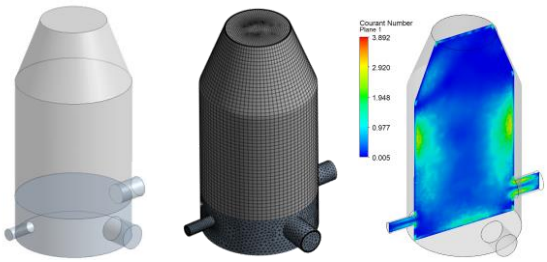
TEM uses a type of pulse machines for thermal pressure treatment utilizing energy generated by the combustion of gas-fuel mixtures. Regarding how the mixture is formed, most studies devoted to the design of systems to generate fuel mixtures for machines of this type have focused on such indicators as drive efficiency, specific effective work and full drive efficiency. Meanwhile, our preliminary research on TEM processing has proven the high significance of and influence on the processing quality of parameters of the fuel mixture such as the mass of the charge, its composition and degree of homogeneity [8]. In turn, the degree of homogeneity depends on a number of filling parameters, including the geometry of the tank being filled [9]. In terms of thermal pulse processing, it is essential that the total amount of heat absorbed per unit surface area of the part during the processing time peaks precisely when the stoichiometric composition of the fuel mixture is achieved. When changing, for example, the mass concentration of the fuel by ±5%, its value increases from 30% (with an excess of oxidizer) to 40% (with an excess of fuel).

Therefore, the subject of this study is the process of mixture formation in a closed combustion chamber using TEM equipment. An experimental setup of a TEM-equipped combustion chamber with a volume of 3.9 liters



a) b)

Figure 1: Experimental setup of TEM-equipped combustion chamber (a) and its CAD model (b)



a) b) c)

Figure 2: The internal cavity of the combustion chamber (a), the computational mesh (b) and the Courant number (c)

(*Figure 1a*) and its CAD model (*Figure 1b*) consists of two specified inlets through which fuel and oxidizer enter. The outlet is used to achieve a vacuum in the chamber before the fuel mixture enters and remove combusted gases afterwards. The inlets through which the fuel and oxidizer enter were separated to accurately measure the amount of fuel gas produced by the electrolyzer.

The calculation domain is the internal cavity of the combustion chamber (*Figure 2a*). The computational mesh (*Figure 2b*) was built in the ANSYS CFX Mesh module to make further calculations in the ANSYS CFX system [10]. The computational mesh, consisting of 182735 finite volumes, combines tetrahedra and hexahedra. To correctly model the near-wall flow of gases, 5 prismatic layers with a growth factor of elements equal to 1.2 were built. The computational mesh was chosen ensuring that the Courant number fell within the range from 2 to 10 to maintain a balance between the accuracy of the obtained results and the performance of the calculations. The Courant number determines the relationship between the timestep and the resolution of the mesh at a given flow rate (*Figure 2c*).

During the filling stage, the accuracy of the composition of the mixture is ensured by using appropriate conditions of needed mass. During the calculations, the Courant number during the holding stage did not exceed 7.8, particularly in the near wall zones which are not critical with regard to the investigated mixing process in the chamber.

Mixing of the hydrogen-air mixture in the expected stoichiometric ratio according to the reaction

$$2H_2+O_2+3.71N_2 = 2H_2O+3.71N_2$$

was investigated. The direction of the subsonic gas flow through the inlets was normal. The calculation was carried out in a transient setting. During the simulation, the value of the molar fraction of H_2 was monitored along the height of the chamber.

The stages over which the mixture formed are shown in *Figure 3*. In the study, the following initial conditions were set: initial pressure after achieving a vacuum was 0.01 MPa, initial temperature of the chamber wall and gas was 20 C. The mass flow rates of H_2 and air were $8 \cdot 10^{-8}$ and $3.137 \cdot 10^{-5}$ kg/s, respectively.

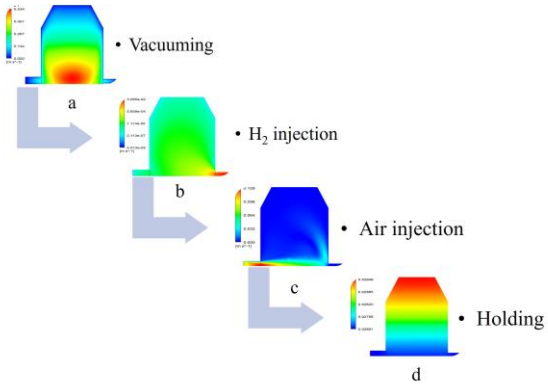


Figure 3: Schematic diagram of the stages over which the mixture was formed: flow velocity contours (a – c) and H_2 volume fraction contour (d)

3. Mathematical model

The gas mixing in the chamber and the gas path elements are described by a system of three-dimensional equations of single-phase multicomponent flow, including the laws of the conservation of mass, momentum and energy in their differential forms. The component balance equation is used to determine the distribution regarding the composition of the mixture. In general, the equations used to describe the generation of a fuel mixture are presented in *Table 1*.

The stress tensor can be represented as the sum of $\boldsymbol{P} = -p\boldsymbol{I} + \boldsymbol{\tau}$, where $\boldsymbol{I}$ denotes the unit tensor and $\boldsymbol{\tau}$ represents the deviatoric stress tensor. The last terms in *Equations 2* and *3* take the forms $\nabla \cdot \boldsymbol{P} = -\nabla p + \nabla \cdot \boldsymbol{\tau}$ and $\boldsymbol{P}: \nabla \mathbf{u} = -p\nabla \cdot \boldsymbol{u} + \boldsymbol{\tau}: \nabla \boldsymbol{u}$, respectively.

The system of *Equations 1-5* is somewhat different with regard to supplying the components of the mixture and storing the fuel mixture in the working chamber due to the features of mixture formation described above. When mixing gases that are fed into the chamber or elements of the gas path, the main factor is the velocity of the jets of gas. Therefore, in *Equations 2* and *3*, the mass forces given by the terms $\sum_{i=1}^{N} \rho^i \boldsymbol{g}$ and $\sum_{i=1}^{N} \boldsymbol{j}^i \boldsymbol{g}$ can be neglected. The rate of thermal and barodiffusion while mixing gases to form the fuel mixture is much lower than that of molecular and turbulent diffusion, allowing these processes to be neglected when writing *Equations 5*. Since the rate of molecular diffusion is negligible compared to that of mechanical gas mixing, molecular diffusion can be ignored when modeling the filling of the working chamber with the gas mixture components and only taken into account while the fuel mixture is being stored in the working chamber.

Table 1: The equations used to describe the generation of a fuel mixture

During the gas supply stage:	The mixture in the chamber during the holding stage:	
$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \boldsymbol{u}) = 0$		(1)
$\rho \frac{D\boldsymbol{u}}{Dt} = \nabla \cdot \boldsymbol{P}$	$\rho \frac{D\boldsymbol{u}}{Dt} = \sum_{i=1}^{N} \rho^i \boldsymbol{g} + \nabla \cdot \boldsymbol{P}$	(2)
$\rho \frac{de}{dt} = \nabla \cdot (\lambda \nabla T) + \boldsymbol{P}: \nabla \boldsymbol{u}$	$\rho \frac{de}{dt} = \nabla \cdot (\lambda \nabla T) + \sum_{i=1}^{N} \boldsymbol{j}^i \cdot \boldsymbol{g} + \boldsymbol{P}: \nabla \boldsymbol{u}$	(3)
$\rho \frac{\partial c^i}{\partial t} = -\nabla \cdot \boldsymbol{j}^i, \quad i = 1, \ldots, N-1$		(4)
	$\boldsymbol{w}^i = -(D^i + D^{iT}) \nabla c^i$	(5)

$\frac{D}{Dt}$: material derivative defined as $\frac{\partial}{\partial t} + \boldsymbol{u} \cdot \nabla$;
$\rho(\rho^i)$, $p(p^i)$: density and pressure of the mixture and its components;
$\boldsymbol{P}$: stress tensor;
c^i : mass concentration of the i-th component of the mixture;
e : internal energy of a gas;
N : number of components in the mixture;
$\boldsymbol{j}^i = \rho^i \boldsymbol{w}^i$: diffusion flux vector;
D^i, D^{iT} : coefficients of molecular and turbulent diffusion of the i-th component in the mixture.

When calculating the density, pressure, enthalpy and gas constant of the mixture, the mixing rules for ideal gases are used:

$$1/\rho = \sum_{i=1}^{N} c_i/\rho_i, \quad p = \sum_{i=1}^{N} p_i,$$
$$\Re = \sum_{i=1}^{N} c_i \Re_i, \qquad H = \sum_{i=1}^{N} c_i H_i \qquad (6).$$

The equations of state and experimental data are used to determine the thermophysical properties of a fluid, i.e. dependence of the density, viscosity, thermal conductivity, specific heat capacity and diffusion coefficients of fluids on the pressure and temperature. Taking into account the fact that the flow in fuel-mixture generation systems of thermo-imPULSE equipment is significantly turbulent, in order to close the system of *Equations 1-6* it is necessary to also use one of the turbulence models integrated into standard computational gas dynamics packages. When choosing a turbulence model and formulating the system of governing equations, the features inherent when generating the fuel mixture in a thermo-imPULSE equipment were taken into account. Considering the requirements to achieve computational efficiency, semi-empirical turbulence models based on the Reynolds-averaged Navier-Stokes (RANS) equations were considered to construct the computational model.

Fourier's law for turbulent heat flow was used to estimate the temperature of the gas:

$$\boldsymbol{q}_{\mathrm{T}} = -\lambda_t \nabla \mathrm{T} \qquad (7),$$

where the turbulent heat transfer coefficient was defined by:

$$\lambda_t = \frac{C_p \mu_t}{Pr_{\mathrm{T}}} \qquad (8),$$

and Pr_{T}denotes the turbulent form of the Prandtl number.

The equations obtained from Reynolds averaging describe the behavior of the averaged characteristics of the turbulent flow, where the viscosity and thermal conductivity coefficients are taken as effective values equal to the sum of the laminar as well as turbulent viscosity and thermal conductivity, respectively, i.e.:

$$\mu = \mu_l + \mu_t, \quad \lambda = \lambda_l + \lambda_t \qquad (9).$$

To simulate turbulence, the Shear Stress Transport (SST) model was used being the most suitable in such cases [11].

The system of *Equations 1-9* fully determines the characteristics of a multi-component gas mixture when mixing while feeding components into the working chamber or elements into the gas flow as well as during aging after the gas has been supplied. As noted in the previous section, based on the specific requirements associated with the peculiarities of thermal energy treatment, the model should be supplemented with criteria for assessing the quality of the fuel mixture.

Such criteria may include:

– the accuracy of its composition

$$\frac{M(c_n)=\int_V c_n dV}{\int_V dV} \in [c_n] \pm \delta(c_n) \qquad (10);$$

– homogeneity of the fuel mixture

$$\sigma(c_n) = \sqrt{\frac{\int_V \left(c_n - \frac{\int_V c_n dV}{\int_V dV}\right)^2 dV}{\int_V dV}} \leq [\sigma(c_n)] \qquad (11);$$

– dosing accuracy of the charge

$$G_{nc} = \int_V \rho dV \in [G_{nc}] \pm \delta(G_{nc}) \qquad (12).$$

The first criterion (*Equation 10*) determines how accurate the composition of the mixture is which is especially important when mixing during thermoplastic pulse processing. An excess of oxidizer in such processes can lead to the ignition of the processed parts and an excess of fuel (if hydrocarbon compounds are used as the fuel) can lead to soot deposits on the processed parts.

The second criterion (*Equation 11*) is the standard deviation of the mass concentration of the fuel in the mixture within a certain volume compared to a set value and is the main parameter when designing mixing devices as well as determining the holding time of the mixture in the working chamber from the moment the gas supply stops until the mixture is ignited.

Finally, the third criterion (*Equation 12*) determines the accuracy with regard to the mass dosing of the charge and is crucial to stabilize the processing quality.

4. The obtained simulation results

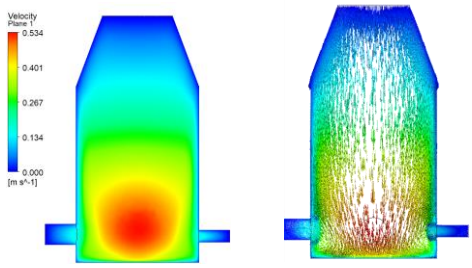
As a result of CFD modeling, the velocity fields of gas flows, components of gas mixtures and volume fractions inside the chamber were determined. The velocity profiles and vector patterns over a cross-section of the chamber at corresponding stages of mixing while the chamber was being filled are shown in *Figures 4-6*.

While establishing a vacuum, the air was pumped out from the chamber until the pressure fell to 0.01 MPa. After that, H_2 and air were sequentially injected inside the chamber until a stoichiometric mixture was achieved.

Injecting components of the mixture this way provided both a high concentration of its components and dosing accuracy of its charge. Accuracy criteria (*Equation 10* and *12*) were used to define the exact mass of H_2 and air corresponding to the stoichiometric mixture, moreover, were used as termination criteria for calculations. Although the accuracy of its composition and dosing accuracy of the charge did not exceed 2%, such heterogeneous flow conditions affected the accuracy of the mixing processes inside the chamber because of the different densities of H_2 and air. The distributions of the molar concentration of H_2 over the characteristic cross-sections of the chamber after injecting air are presented in *Figure 7*.

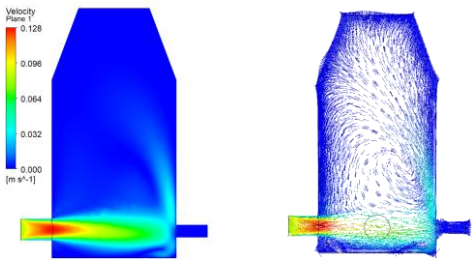
The results show that the normalized standard deviation of the H_2 molar fraction (10) was equal to 20%. The mixture was poorly homogeneous and needed to be held.

Therefore, after injecting the components, the mixture was held. Several monitoring points were used to control the H_2 volume fraction in the chamber at different heights (*Figure 8a*).



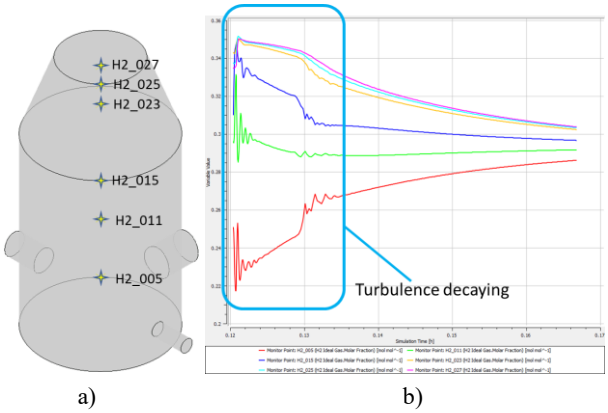
a) b)

Figure 4: Gas flow field in the central section of the chamber while establishing a *vacuum* (t=0.73 s after the process was started)
(a) velocity profile; (b) vector patterns



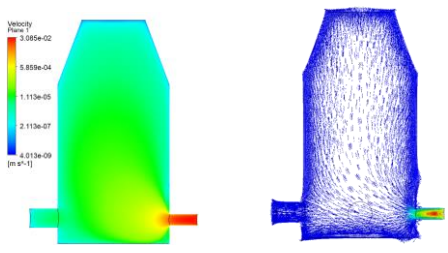
a) b)

Figure 6: Gas flow field in the central section of the chamber while *injecting air* (t=600 s after the process was started)
(a) velocity profile; (b) vector patterns



a) b)

Figure 8: H_2 volume fraction at different heights in the chamber
(a) location of the monitoring points;
(b) H_2 volume fraction vs. holding time



a) b)

Figure 5: Gas flow field in the central section of the chamber while *injecting* H_2 (t=433.23 s after the process was started)
(a) velocity profile; (b) vector patterns

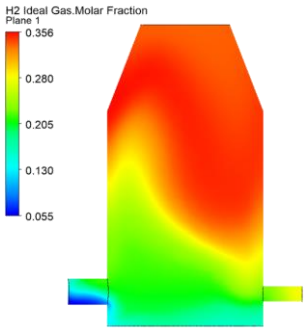


Figure 7: H_2 molar fraction in the central section of the chamber after injecting air (t=600 s after the process was started

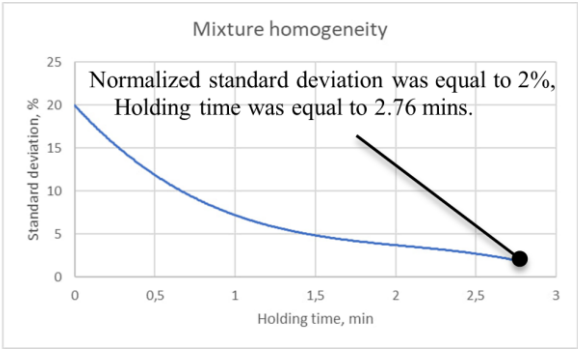


Figure 9: Homogeneity of the mixture vs. holding time

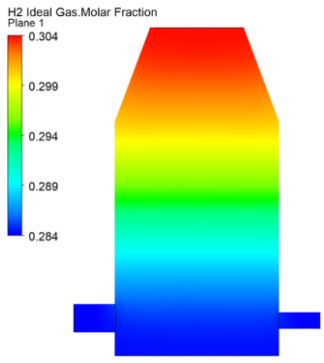


Figure 10: H_2 molar fraction in the central section of the chamber after being held (t=766 s after the process was started

After holding the mixture for 1 minute, the gas stopped moving and turbulence decay was observed. During approximately the following 2 minutes, the fuel mixture slowly became more homogeneous as a result of diffusion (*Figure 8b)* until the accuracy of homogeneity reached 2% (*Figure 9)*:

The final distribution of the H_2 volume fraction after being held for approximately 3 minutes is shown in *Figure 10*.

5. Conclusions

CFD simulation results showed that the mixer provides a high degree of homogeneity of the fuel mixture. According to the normalized estimate of the root mean square deviation of the H_2 molar concentration, the deviation of the methane content from the stoichiometric value did not exceed 2%.

According to calculations, the mixture needed to be held for approximately 3 minutes to achieve the required homogeneity of the gas mixture in the combustion chamber, which was longer than expected. This problem can be solved by modernizing the main inlets of the components as well as introducing additional mixing mechanisms. However, changes in the configuration of TEM equipment will be required while implementing the pilot technology during cyclic production. For research purposes, the holding time does not affect the essence of the process but must be determined and taken into account.

In addition, the influence of heating the walls of the combustion chamber during its cyclic operation on the quality indicators of the fuel mixture needs to be further studied.

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