The study of thermostressed condition of the hydrogen quartz reactor during the methane pyrolysis

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Abstract. Based on analytical and numerical methods, the detailed studies of the temperature and thermostressed condition of quartz reactor intended for production of hydrogen by pyrolysis of methane passed through the molten tin, are performed. Heating of tin contained in the reactor is realized by the application of electromagnetic field generated by the inductor. The analysis of the study results allows concluding that the values of radial, circular and axial stresses during a uniform movement of the reactor along its height even in the most unfavourable conditions when the temperature drop along the wall of the quartz reactor is maximum and is equal to the difference of temperatures between molten tin and environment \((\Delta T \approx 1500^\circ C)\) are within the tensile strength limit for quartz \(\sigma_p = 50 Mpa\) (5 kg / mm\(^2\)). In case of non-uniform heating of the reactor along its height observed at its partial filling with molten tin, in the area of transfer from the filled part of the reactor to its non-filled part, there are high axial temperature gradients, which result in temperature stresses exceeding the tensile strength limit. By using the obtained results, the recommendations connected with the reactor heating rate and essential for ensuring its safe operation have been developed.

1. Determination of quasistatic temperature stresses in a reactor crucible

1.1. Analytical calculation of temperature stresses in the reactor bowl

By using the data on wall temperature condition of quartz reactor, we found the radial, circular and axial quasistatic temperature stresses \(\sigma_r, \sigma_\phi, \sigma_z\). The formulas for their calculation in case when the temperature on inner cylinder surface is equal to \(T = 1500^\circ C\), and on the outer \(T = T_0 = 0^\circ C\), have the form of \([1 – 3]\)

\[
\sigma_r(r) = \frac{aET}{2(1-\nu)\ln \frac{r_2}{r_1}} \left[ -\ln \frac{r_2}{r} - \frac{r_1^2}{r_2^2 - r_1^2} \left( 1 - \frac{r_1^2}{r_2^2} \right) \ln \frac{r_2}{r_1} \right]; \tag{1}
\]

\[
\sigma_\phi(r) = \frac{aET}{2(1-\nu)\ln \frac{r_2}{r_1}} \left[ -\ln \frac{r_2}{r} - \frac{r_1^2}{r_2^2 - r_1^2} \left( 1 + \frac{r_1^2}{r_2^2} \right) \ln \frac{r_2}{r_1} \right]; \tag{2}
\]
\[ \sigma_r(r) = \frac{a E T}{2(1-\nu) \ln \frac{r_2}{r_1}} \left[ 1 - 2 \ln \frac{r_2}{r} - \frac{2r_1^2}{r_2^2 - r_1^2} \ln \frac{r_2}{r_1} \right], \]

where \( a \) is the temperature linear expansion factor; \( E \) is the elasticity modulus; \( \nu \) is the Poisson coefficient; and \( r_1, r_2 \) are the inner and outer radius of hollow cylinder (figure 4).

It should be noted that with formulas (1) – (3) quasistatic temperature stresses were found. The values of \( a, E, \nu, r_1, r_2 \) for quarts reactor were assumed as follows: \( a = 0.55 \cdot 10^{-6} 1/ K; E = 73 \cdot 10^8 Pa; \nu = 0.17; r_1 = 0.0075 \text{m}; r_2 = 0.0105 \text{m}. \)

1.2. The analysis of temperature stress calculation results

The results of temperature stress calculations by the formulas (1) – (3) are given in figures 1 - 3. It follows from their analysis that radial temperature stresses \( \sigma_r \) on the inner and outer surfaces of the reactor take zero values (figure 1). They reach the maximum value \( \sigma_r = 3.6 \text{MPa} (0.36 \text{kg/mm}^2) \) in the wall centre. The negative sign means that the stresses are compressive. Thus, the ultimate compressive strength for quarts amounts to \( \sigma_p = 1100 \text{MPa} (110 \text{kg/mm}^2) \), as a result the value of these stresses is safe for the reactor operation.

Distribution of circular temperature stresses within the reactor thickness is given in (figure 2). It follows from their analysis that the maximum compressive stresses \( \sigma_{\sigma_0} (r_1) = 48 \text{MPa} (4.8 \text{kg/mm}^2) \) occur at the inner (more heated) surface of the reactor. Within the wall thickness, the stresses \( \sigma_\sigma \) change their sign and tension stresses are observed at the outer (less heated) surface, which amount to \( \sigma_{\sigma_0} (r_2) = 39 \text{MPa} (3.9 \text{kg/mm}^2) \). It should be noted that the value of these stresses is at the strength limit for the given material, amounting to \( \sigma_{\sigma_0} (r_1) = 50 \text{MPa} (5 \text{kg/mm}^2) \). The distribution of axial stresses \( \sigma_z(r) \) insignificantly differs from the stresses \( \sigma_{\sigma_0} (r) \) (figure 3).

![Figure 1. The distribution of radial temperature stresses along the reactor wall thickness.](image-url)
Figure 2. Distribution of circular temperature stresses along the reactor wall thickness.

Figure 3. Distribution of axial temperature stresses along the reactor bowl.

It follows from the results of calculations of radial $\sigma_r(r)$, circular $\sigma_\varphi(r)$ and axial $\sigma_z(r)$ stresses that the stresses $\sigma_\varphi(r_2)$ and $\sigma_z(r_2)$ on the outer surface reach ultimate values for the tension stress. However, it should be noted that the stresses were calculated for the most unfavourable temperature
conditions when the temperature drop along the wall thickness was maximum and amounted to $\Delta T = 1500^\circ C$. Under real conditions, the measures to increase the value of $\Delta T$ should be taken.

It should be noted that the given analytical ratios (1) - (3) for determining radial, circular and axial temperature stresses do not consider the temperature gradients along the reactor bowl height. In this connection, the given work describes the numeric simulation of temperature and thermoelastic problems in a three-dimensional setting by the finite element method in the Transient Thermal and Structural modules of the Ansys 18.2 software complex.

2. Thermal and strength calculation of the reactor bowl

2.1. Mathematical problem setting
The analytical calculations for temperature fields using precision [4, 5] and approximate [6,7] methods as well as the analytic calculation of temperature stresses [8] in case of considering the variable heat transfer coefficient along the reactor height and heat exchange by radiation in the gas medium are difficult. Therefore, for determining the temperature fields and thermal stresses the finite element method is used.

Let us consider the boundary heat conductivity problem taking into account a complex exchange in the gas layer for the quarts reactor (hollow finite cylinder) in the following mathematical setting (figure 4)

$$\frac{\partial T(r,z,t)}{\partial t} = \alpha \left( \frac{\partial^2 T(r,z,t)}{\partial r^2} + \frac{1}{r} \frac{\partial T(r,z,t)}{\partial r} + \frac{\partial^2 T(r,z,t)}{\partial z^2} \right),$$

\[T(r,z,0) = T_0;\]

\[\frac{\partial T(r,z,t)}{\partial r} = 0, \quad (0 < z < z_2);\]

\[\lambda \frac{\partial T(r,z,t)}{\partial r} + \alpha_1 [T_{ml} - T(r,z,t)] = 0, \quad (0 < z < z_1);\]

\[\lambda \frac{\partial T(r,z,t)}{\partial r} + \alpha_2 [T_{m2} - T(r,z,t)] + e \left[ \frac{T_{m2}^4}{100} - \frac{T(r,z,t)^4}{100} \right] = 0, \quad (z_1 < z < z_2);\]

$$\frac{\partial T(r,0,t)}{\partial z} = 0, \quad (\frac{r_1}{2} < r < r_2);$$

$$\frac{\partial T(r,z_2,t)}{\partial z} = 0, \quad (r_1 < r < r_2);$$

where $T$ is the temperature; $r$, $z$ are the radial and axial coordinates; $t$ is the time; $\alpha$ is the temperature conductivity coefficient; $\lambda$ is the thermal conductivity coefficient; $\alpha_1$, $\alpha_2$ are the heat transfer coefficients of the liquid and gas medium; $T_{ml}$, $T_{m2}$ are the medium temperatures; $T_0$ is the initial bowl temperature; and $e_r$ is the reduced emissivity factor of the radiator system $C_0 = 5.67 \frac{W}{m^2 K}$ – Stephan - Boltzmann constant.
2.2. The numerical solving and calculation result analysis

The problem (4) – (10) was solved by the numerical method [10]. The source data for solving the problem were as follows: \( r_1 = 7.5\, \text{mm} \); \( r_2 = 10.5\, \text{mm} \); \( z_1 = 0.37\, \text{m} \); \( z_2 = 0.5\, \text{m} \); \( \alpha_1 = 20000\, \text{W}/(\text{m}^2\, \text{K}) \); \( \alpha_2 = 100\, \text{W}/(\text{m}^2\, \text{K}) \); \( T_{m1} = 1500^\circ\text{C} \); \( T_{m2} = 1000^\circ\text{C} \); and \( T_0 = 0^\circ\text{C} \).

From the mathematical setting of the problem, it follows that with the maximum power of the induction heating, the temperature of the skin layer of the molten metal at the boundary with the reactor wall at the initial moment of time becomes equal to \( T_{m1} = 1500^\circ\text{C} \) (at the section \( 0 < z < z_1 \)). The medium temperature at the section \( z_1 < z < z_2 \) was assumed to be equal to the temperature of methane hydrogen mixture \( T_{m2} = 1000^\circ\text{C} \) obtained after passage of methane through the layer of the molten metal. It should be noted that in the contact zone of the molten metal and inner surface of the bowl \( 0 < z < z_1 \), the heat exchange was simulated using the Newton-Richmann law (7), and in the contact zone of gas mixture and inner surface of the bowl at a height of \( z_1 < z < z_2 \) by complex heat exchange laws (8), i. e. the simultaneous flowing of the convection and radiation was considered in the computer model. This approach allowed developing the most reliable computer model of heat exchange processes in the reactor.

The results of calculations for the problem (4) – (10) are given in figs. 5 – 10. Figure 5 shows the temperature distribution in the bowl at a contact level of the gas and liquid media. The finite element grid in the contact location is condensed in view of the fact that heat transfer coefficient for the liquid medium is 200 times higher as compared to the gas one, and a sharp temperature jump (figure 5, 6) is observed with the temperature difference of \( \Delta T = 1320^\circ\text{C} \) at the time moment \( t = 1\, \text{s} \).
Figure 5. Finite element grid and temperature distribution in the bowl at a contact level of the gas and liquid media.

Figure 6. Temperature distribution along the bowl height.

Figure 7 shows temperature distribution along the bowl thickness at the liquid medium level. It follows from its analysis that the maximum temperature drop along the reactor wall in the area of molten metal is observed at the moment of time $t = 1\, s$, and amounts to $\Delta T = 1330^\circ C$ (figure 7). This is explained by the fact that due to high coefficient of heat transfer from the molten metal to the
reactor wall $\alpha = 20000 \text{ W} / (\text{m}^2\text{K})$, its temperature at the boundary takes the melt temperature almost immediately. Thus, in this case the heat exchange conditions are assumed; they result in the maximum temperature gradients in the reactor wall, which ensure its operational safety margin according to the thermal strength conditions.

![Temperature distribution along the bowl thickness at the liquid medium level](chart.png)

**Figure 7.** Temperature distribution along the bowl thickness at the liquid medium level: $\delta$ - reactor wall thickness.

Figures 8, 9 give the distribution of equivalent temperature stresses according to Mises in the cross-section at the level of liquid metal (figure 8) and gas (figure 9) media in time. It should be noted that the value of temperature stresses at the level of liquid metal medium is one order of magnitude higher than the stress in the bowl at the gas medium level. Figure 10 shows the distribution of temperature stresses along the bowl height in time. Here a great difference between the stresses in the reactor bowl parts filled with liquid metal $0 < z < 0.33 \text{ m}$ and gas $0.33 \text{ m} < z < 0.5 \text{ m}$ medium may be seen. At the interface of two media $z = 0.33 \text{ m}$ a jump of temperature stresses increasing in time can be observed, which, at $t = 1 \text{ s}$ reaches the values exceeding the strength limit for the quarts $\sigma_s > 50 \text{ MPa}$ ($5 \text{ kg} / \text{ mm}^2$).
Figure 8. The distribution of equivalent temperature stresses according to Mises in the cross-section of the bowl at a level of liquid metal medium in time.

Figure 9. The distribution of equivalent temperature stresses according to Mises in the cross-section of the bowl at a level of gas metal medium in time.
Figure 10. The distribution of equivalent temperature stresses according to Mises along the bowl height in time.

Figure 11 shows the reactor bowl deformations at the interface of two media as a result of high temperature gradients. A part of the bowl filled with the liquid metal expands considerably more than the upper part filled with the gas, which may finally result in the cracks and failure of the laboratory bench.

Figure 11. The deformation pattern at the interface of two media in the reactor bowl.
Conclusions

1. Based on the data of temperature condition of the quartz reactor, radial, circular and axial quasistatic temperature stresses have been found. Their analysis shows that circular and axial tension stresses observed at the outer surface of the reactor reach the value of 39 MPa (3.9 kg/mm²), located at the strength limit point for the given material $\sigma_p = 50$ MPa (5 kg/mm²).

2. A sharp wall temperature jump is observed at the interface of liquid metal and gas media in the upper part of the reactor in the vertical direction, which is connected with a great difference between the heat transfer coefficients observed during heat transfer from the liquid metal and gas media to the reactor wall. Temperature stresses in this zone found by the Mises theory, exceed the strength limit for the given material.

3. To ensure safe operation of the reactor, a gradual and uniform heating of the bowl shall be performed. The stationary operating mode shall be achieved within 20 minutes to 1 hour by recording the temperatures of the inner bowl surface at the levels of $z = 0.2$ and $z = 0.4$ m during heating. The rate of change of the recorded temperature should not exceed 200 °C/min, furthermore the difference between the readings on the inner bowl surface in the liquid and gas medium should not exceed $\Delta t = 500^\circ C$.

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