MODELING OF THE CROSSING POINT TEMPERATURE PHENOMENON IN THE LOW-TEMPERATURE OXIDATION OF COAL

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ABSTRACT

In this study, modeling of the crossing point temperature (CPT) phenomenon in the low-temperature oxidation of coal was carried out using COMSOL Multiphysics®. Low-temperature oxidation can lead to spontaneous combustion of coal stockpiles. The CPT phenomenon was modeled with the kinetics data obtained from a prior laboratory experimental study. The coupling of the heat-transfer phenomenon through conduction and convection determined the thermal evolution model. In this case, coal received the initial heat of the oven temperature increases. As the coal temperature rose, the heat generated from oxidation was released into the environment via conduction and convection. Meanwhile, oxidation products and oxygen were transferred by convection and diffusion. The effects of moisture and the humidity were not considered. The outcomes of modeling were validated through comparison with the results of experimental tests, and the modeling result agreed well with the experiment tests, with temperature deviations of about 0.9%. The effects of airflow rate, oxygen concentration, porosity, and the initial temperature on low-temperature coal oxidation were also examined.

Keywords: Coal; Crossing point; Low-temperature oxidation; Modeling; Spontaneous combustion

1. INTRODUCTION

Research and publications on the spontaneous combustion of coal have been reported by several researchers (Carras & Young, 1994; Nugroho et al., 1998; Sujanti et al., 1999; Nugroho et al., 2008; Saleh & Nugroho, 2013). Low-temperature oxidation leading to coal spontaneous combustion takes place when the heat generation rate of the coal pile exceeds the rate of heat dissipation to the environment. It is important for sufficient heat to be supplied or sufficient oxygen to be available to trigger the exothermic chemical reaction of coal and oxygen. The reaction rate of coal oxidation will increase with increasing coal temperature, and the reaction is irreversible.

One method that is widely applied to study spontaneous combustion is the crossing point temperature (CPT) method. The CPT method applies the temperature at which the coal temperature starts to exceed the temperature of the surroundings (the oven temperature). This approach was first implemented using the oil-bath method by Nubling and Wanner (1915), who

used a constant rate of heating to evaluate the coal CPT. A new testing method was proposed by Chen and Chong (1998), who utilized a cubic or chamber basket to determine CPT. Following this, other researchers utilized Chen and Chong's (1998) method (Nugroho et al., 1998; Zhong et al., 2006). They studied spontaneous the combustion behavior of coals under diverse test conditions. Furthermore, Xuyao et al. (2011) applied the CPT testing approach using a plug flow reaction vessel.

In an investigation of the safety of coal during storage and transport, it is desirable to model the spontaneous combustion phenomenon in greater detail based on their physical and chemistry properties (Salinger et al., 1994; Krishnaswamy et al., 1996; Akgun & Essenhigh, 2001; Yuan & Smith, 2008). This paper aims to model the CPT phenomenon in low temperature oxidation of coal using COMSOL Multiphysics®. This model enables the coupling of thermodynamics, heat transfer, chemistry, and physical properties of the coal bed into the geometry of the system. The outcomes of the simulation are compared with experimental data.

2.   CROSSING POINT TESTS

The experimental study of the CPT method was performed using South Banko coal with the experimental setup following that given by Xuyao et al. (2011). Figure 1 shows the CPT testing method, which comprises an apparatus to simulate the coal oxidation reaction and a temperature recording instrument. The simulation apparatus consists of an oven as an enclosed space where the temperature can be controlled and programmed, a coal reaction vessel, and a gas cylinder.

The oven serves as a gas bath that provides a circumambient surrounding at an appropriate and equal temperature. The temperature of dry air is equal to the circumambient temperature, since the dry air passes through the preheating tube prior to entering the coal reaction vessel. Pure copper is used as material for the manufacture of coal reaction vessels due to its excellent thermal conductivity. Figure 1b shows that at the bottom, there is a space as high as 20 mm separated by a stainless-steel wire mesh size of 0.15 mm as a gas buffer to ensure a steady flow of dry air in the coal sample. Here, thermocouple 1 was used to monitor the ambient temperature arranged in the middle of the oven. Meanwhile, thermocouple 2 was used to monitor the coal temperature, and it was placed in the middle of the coal reaction vessel.
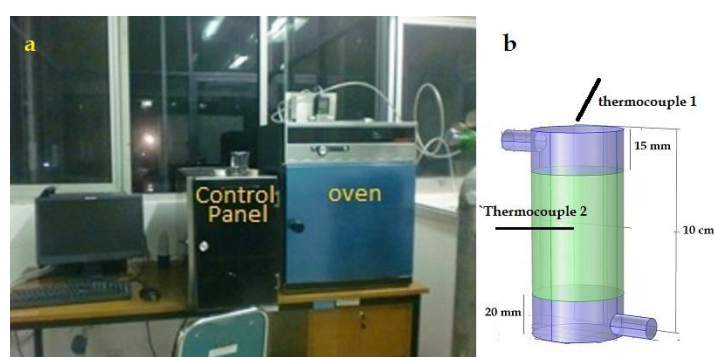

Figure 1 Photo of the crossing-point testing (CPT) apparatus

The experiment started with filling the coal reaction vessel with 50 g of South Banko coal. Furthermore, the oven temperature was programmed to operate at a fixed temperature of 40°C, and 0.2 L/min of dry air with a 21% concentration of oxygen was allowed to stream into the coal reaction vessel. Coal and ambient temperatures were observed continuously using the temperature recording instrument. The oven was regulated to escalate the temperature at a scheduled rate of 1°C/min when the coal reached a temperature of 40°C, while the dry airflow rate of 0.2 L/min was retained. The test ended when the temperature of coal was 5°C above the

ambient temperature. The results of the experimental works are compared with the modeling results in the next section 3.

2.1. Modeling of Low-temperature Coal Oxidation

The oxidation of coal at low temperatures represents a complicated reaction. Three mechanisms commonly expected to occur are as follows (Carras & Young, 1994): (i) physical adsorption; (ii) chemical adsorption, leading to the establishment of the complexes of coal–oxygen and carbon–oxygen; and (iii) oxidation, where oxygen oxidizes coal by releasing gaseous products, typically carbon monoxide (CO), carbon dioxide (CO_2), and water vapor (H_2O). Oxidation is the most exothermic reaction of the three processes described above. In daily practice, a major fire hazard due to rapid combustion should be avoided when possible. When rapid coal oxidation occurs, the coal temperature will rise rapidly. The CPT is defined as the temperature at the point where the ambient temperature is exceeded by the temperature of the coal. The propensity of spontaneous combustion can be evaluated from the CPT values.

A good understanding of the complexity of the chemical reaction between coal and oxygen at low temperatures has not been reached. During coal oxidation, the gas products that emerge are mainly CO_2, CO, and H_2O. It has been reported that the oxidation of coal also produces a small amount of a mixture of aromatic acids, oxalic acid, and unsaturated hydrocarbons, such as C_2H_2 and C_2H_4 (Smith et al., 1991). The simulations adopted that assumption that the products of the coal oxidation reaction are only CO_2 and CO. The origin and rank of coal affects its detailed chemical structure. Measurement of the relationship between the amount of oxygen consumed and the level of gaseous production remains complicated. Smith et al. (1991) reported that one mole of oxygen needed by one mole of coal to generate about 0.1 mole of carbon monoxide (CO) and one mole of carbon dioxide (CO_2) plus heat in the early stages of the oxidation of coal. Thus, the equation of the chemical reaction can be written as

$$coal + O_2 \rightarrow CO_2 + 0.1CO + heat \tag{1}$$

The dependence of the oxidation rate, r, on the concentration of oxygen and temperature can be determined by the Arrhenius equation:

$$r = A[O_2]^n \exp(-E/RT) \tag{2}$$

wherein the chemical reaction rate is described as the conversion rate of the reactant and product concentrations, in units of $kmol/(m^3 s)$; A is the pre-exponential factor, in units of $(kmol/m^3)^{1-n} S^{-1}$; E is the energy of apparent activation, in units of kJ/mol; R is the constant of gas, in units of kJ/(mol K); n is the order of reaction; T is the absolute temperature in K; and $[O_2]$ is the oxygen concentration in $kmol/m^3$. The value of the order of reaction, n, for coal and other carbonaceous materials in study of low-temperature oxidation has been proven to differ between 0.5 and 1.0 (Carras & Young, 1994), and for some U.S. coals, it is about 0.61 (Schmidt & Elder, 1940). With this amount, the equation of the rate of reaction will be

$$r = A[O_2]^{0.61} \exp(-E/RT) \tag{3}$$

The apparent activation energy value, E, may differ from 12 to 95 kJ/mol for different coals. The pre-exponential factor, A, relies more on the method of measurement and coal rank, with a typically value among 1 and 7×105/s. The activation energy value and pre-exponential factor for South Banko coal applied in this simulation were obtained by Nugroho et al. (1998) using a basket crossing point method. In their research, the pre-exponential factor and energy of activation obtained by applying the Arrhenius equation were

$$\frac{dT}{dt} = A^* \exp\left(-\frac{E}{RT}\right) \tag{4}$$

$$\rho_s C_{ps} \frac{\partial T}{\partial t} = \rho_s A^* Q \exp\left(-\frac{E}{RT}\right) \tag{5}$$

In conditions where a flat temperature profile exists locally, the rise of temperature is at that instant being balanced approximately by the Newton cooling term at the surface. It follows that when this condition persists, the temperature at the center T_c is

$$\ln\left|\frac{dT}{dt}\right|_{Tc} = \ln\left(\frac{QA^*}{C_{ps}}\right) - \left(\frac{E}{RT_c}\right) \tag{6}$$

Plotting $(1/T_c)$ versus $\ln[dT/dt]_{Tc}$ for different oven temperatures yields an activation energy E of 95 kJ/mol and pre-exponential factor QA^* of 1.84×10^{10} J/ (kg.s) from the slope of an intercept (Nugroho et al., 1998). This equation suggests a reaction rate with a zero order. To distinguish the pre-exponential factor A in Equations 1 and 3, A^* with a unit of K/s was applied to signify the pre-exponential factor for a zero-order reaction. The correlation between an A^* and A may be determined by an implementing the equation for the energy balance in particles of coal:

$$\rho_s C_{ps} \frac{dT}{dt} = Q[O_2]^{0.61} A \exp\left(-\frac{E}{RT}\right) \tag{7}$$

where Q constitutes heat release through the oxidation of coal per mole of oxygen consumed in kJ/mol-O_2, C_{ps} is the coal-specific heat in J/(kg.K), and ρ_s is the density of the coal particle in kg/m^3. Replacing Equation 5 with Equation 7,

$$\rho_s C_{ps} A^* \exp\left(-\frac{E}{RT}\right) = Q[O_2]^{0.61} A \exp\left(-\frac{E}{RT}\right) \tag{8}$$

Hence,

$$A = \frac{\rho_s C_{ps}}{Q[O_2]^{0.61}} A^* \tag{9}$$

As a product of the oxidation reaction, heat is released into the environment by convection and conduction, whereas gaseous products are transferred by diffusion and convection. The process that occurs in the initial phase of self-heating is slow, and thermal equilibrium is assumed to emerge between particles of gas and coal. A detailed heat-transfer-modeling approach applied in this work was available in the literature (Yuan & Smith, 2008).

2.2. Coal Properties and Coal-pile Permeability

Table 1 shows the coal kinetic and physical properties used in the simulation and testing of CPT. In this simulation, the coal pile in the reactor vessel was managed as a porous medium, and the permeability and porosity of the coal pile were used as inputs. The assumptions that coal particles were uniformly packed and had a mean diameter of 0.24 mm were adopted to illustrate the coal pile. The coal-pile porosity was calculated using the following equation:

$$\varepsilon = 1 - \frac{\rho_b}{\rho_p} \tag{10}$$

where ρ_b is the bulk density of coal, and ρ_p is its particle density. Using the respective density of particles and the bulk density of coal, which were 1,260 and 880 kg/m^3, respectively, the porosity calculation result was 0.3. The permeability of the coal pile in the reaction vessel was assumed to be uniform and isotropic. The Kozeny-Carman equations for the stream in packed beds were used to estimate the coal pile permeability (Bird et al., 1966):

$$k = \frac{\varepsilon^3 d^2}{150(1-\varepsilon)^2} \tag{11}$$

where d is the diameter of particles. Applying $\varepsilon = 0.3$ and $d = 2.4e^{-4}$ m, $k = 2.11 \times 10^{-11}$ m^2. The permeability and porosity were assumed to remain stable in the entire oxidation reaction of coal.

Table 1 The kinetic and physical properties of the coal as model inputs for simulation

Parameter	Value	Unit
Bulk density of coal[*]	880	Kg/m^3
Particle density of coal[*]	1,260	Kg/m^3
Conductivity of coal	0.1	W/m.K
Specific heat of coal	1,100	J/kg.K
Heat of reaction	350	kJ/mol-O$_2$
Activation energy[*]	95	kJ/mol
Pre-exponential factor[*]	1.67×10^7	K/s
Initial coal temperature	313.15	K
Coal particle diameter	0.24	Mm

[*]Nugroho et al., 1998

2.3. Numerical Modeling

COMSOL Multiphysics software was applied in this investigation to simulate the gas stream and self-heating in the coal pile reactor (porous media), as well as the gas stream in free media. COMSOL can model the transport and mixing of chemical species by completing the conservation equations representing the reaction, diffusion, and convection component sources for each species. In this study, the gas flow in the reactor vessel was managed as laminar flow in porous media. Because the diameter of fine particles of coal is quite small (the average diameter is about 0.24 mm), the characteristics of the gas stream inside the pile of coal fine-particles can be represented using Darcy's law (Brooks et al., 1988; Salinger et al., 1994; Krishnaswamy et al., 1996; Moghtaderi et al., 2000; Akgun & Essenhigh, 2001). The spontaneous combustion of coal was simulated as a reaction of surface chemicals in which the oxidation reaction appeared on the surface of coal in the porous media. Heat as a product of the oxidation reaction is released into the environment by conduction and convection, whereas gaseous products are transferred by diffusion and convection.

The mass and energy conservation equations leading to the mechanism of oxygen concentrations and heat transport in the reaction vessel of coal are described below. Energy conservation:

$$\left[\varepsilon \rho_g C_{pg} + (1-\varepsilon)\rho_s C_{ps} \right] \frac{\partial T}{\partial t} + \rho_g C_{pg} \left(\frac{\partial(uT)}{\partial x} + \frac{\partial(uT)}{\partial y} + \frac{\partial(uT)}{\partial z} \right) = \lambda_{eff} \left(\frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} + \frac{\partial^2 T}{\partial z^2} \right) + (1-\varepsilon)r\Delta H \tag{12}$$

where ε is the porosity, ρ_g and C_{pg} are the gas density and gas specific heat, ρ_s and C_{ps} are the coal density and coal specific heat, r is the oxidation rate calculated using Equation 2, Q is the reaction heat for oxidation of coal, and λ_{eff} is the effective coal matrix thermal conductivity. This can be rewritten as:

$$\lambda_{eff} = \varepsilon\lambda_g + (1-\varepsilon)\lambda_s \tag{13}$$

where λ_g and λ_s are the thermal conductivities for gas and coal, respectively.

The equation for transferring the concentration of oxygen and the oxidation product is given as follows:

$$\varepsilon\frac{\partial C_i}{\partial t} + \left(\frac{\partial(uC_i)}{\partial x} + \frac{\partial(vC_i)}{\partial y} + \frac{\partial(wC_i)}{\partial z}\right) = D\left(\frac{\partial^2 C_i}{\partial x^2} + \frac{\partial^2 C_i}{\partial y^2} + \frac{\partial^2 C_i}{\partial z^2}\right) + (1-\varepsilon)r \tag{14}$$

where C_i is the concentration of the gas component; u, v, and w are the velocities for the x, y, and z directions, respectively; and D is the diffusion coefficient.

The input data for the simulation are the boundary and initial conditions. As the initial conditions of both coal and air involved a temperature of 313.15 K, the flow applied for CPT experiments was also used as the boundary condition in this simulation, as presented in section 2.2.

The simulation was carried out in two steps. The first was a stationary step without the oxidation reaction of coal to obtain a steady-state flow field and a stable distribution of gas in the coal pile in the reactor vessel and the free regions. The second step involved time-dependent calculations, including the oxidation reaction of coal, and the time step was 1 s. Figure 2 shows the geometrical layout and mesh applied for this simulation. The element size was 0.15 mm, while the total domain element number was about 84,374.
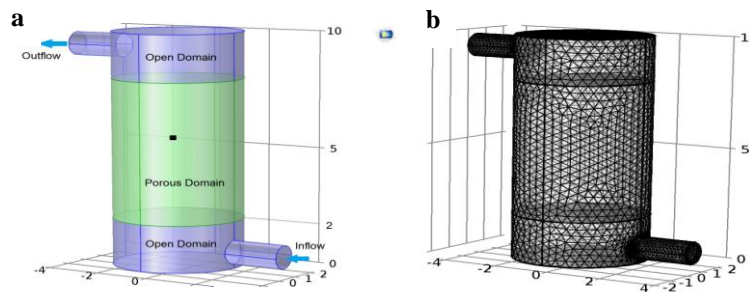

Figure 2 The geometrical layout for: (a) the reactor vessel; and (b) mesh used in the simulation

3. RESULTS AND DISCUSSION

First, the simulation result was validated in comparison with the experimental data. Then, a sum of simulations was performed to assess the influence of four factors (i.e., initial temperature, porosity, concentration of oxygen, and gas flow rate) on the properties of coal spontaneous combustion and CPTs.

Figures 3–4 show the results of the simulation, which demonstrated a streamlined flow and distribution of velocity throughout the reaction vessel. The gas flow rate decreased significantly when entering the porous media and was distributed evenly throughout the coal pile, which
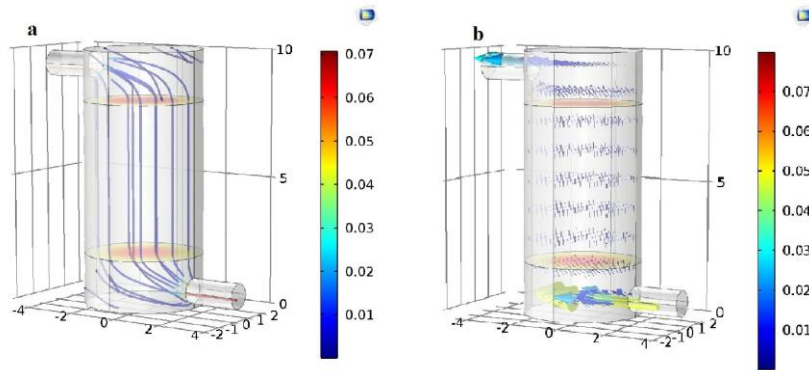
indicates that oxygen may also be distributed evenly.

Figure 3 Streamline (a) and velocity vector (b) (21 vol. % O_2 at initial temperature 313.15 K with a gas flow of 0.2 L/min)

Figure 4 shows the evolution of the heating process in a coal pile, which can be segregated into two phases. Throughout the first phase of spontaneous heating, the coal pile temperature increases; this is mainly due to the rise in oven temperature. In the second phase, due to rising temperatures in the coal pile, the coal oxidation rate also increases and contributes to the increase in the rate of temperature rise until the CPT is reached. These results also indicate that the hotspot was first established at the bottom of the coal pile, and when the CPT was reached, the bottom temperature was about 10 degrees higher. This needs to be taken into consideration for the development of this method, particularly in terms of thermocouple placement.
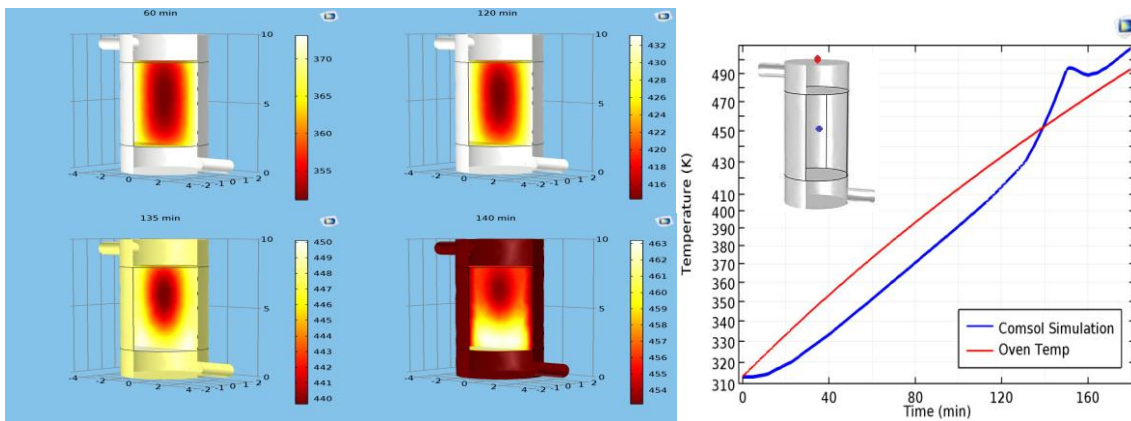

Figure 4 Temperature profiles (21 vol. % O_2 at an initial temperature of 313.15 K with a gas flow of 0.2 L/min)

Figure 5 shows the comparison between the simulation results and experiment data. From Figure 5, it is clear that the temperature predicted by the simulation is in good congruence with the experimental outcomes. At the initial stages to about 350 K, it seems that the experimental data show higher results than the simulation; this is likely because South Banko coal contains enough volatile matter to be sufficiently reactive at low temperatures.

3.1. Effect of the Initial Temperature

The simulation results concerning the effect of initial temperature on CPT is summarized in Figure 6. These results indicated that the CPT exhibits a decline of about 15 degrees with the initial temperature increasing from 30 to 40°C, but it is almost constant with the initial temperature increasing from 40 to 60°C. In contrast, the time to reach the CPT decreased by about 10 minutes with a 10-degrees decline from the initial time.
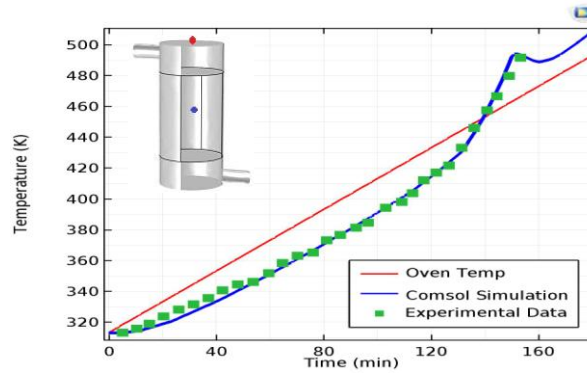
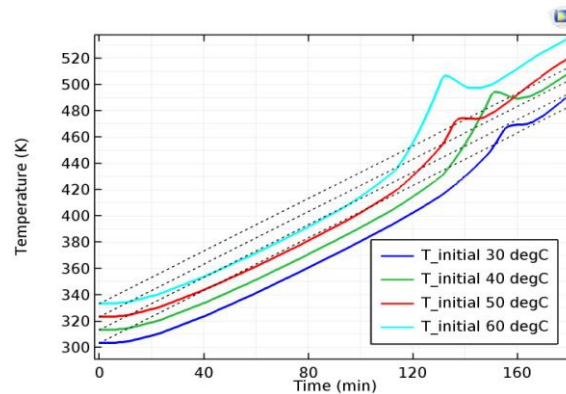Figure 5 Comparison between the simulation and experimental results

Figure 6 Temperature evolution at 21 vol. % O_2 with a 0.2 L/min gas flow at various initial temperatures

3.2. Effect of Porosity

It can be seen in Figure 7 that larger coal-bed porosity results in a faster temperature rise and shorter CPT period. This is because porosity influences the resistance of the airflow. The larger the porosity, the smaller the resistance of the airflow, which enhances the oxidation reaction process. However, larger porosity can also accelerate heat convection, and thus it contributes to the self-heating of coal (Ejlali et al., 2012).

Figure 7 shows the influence of porosity; it illustrates that if the porosity decreases by 10%, CPT will increase by approximately 5 degrees and the time to reach the CPT decreases by about 10 min. Commonly, the coal pile porosity relates to heat and oxygen transports (Akgun & Essenhigh, 2001). Nevertheless, with the flow of gas in the coal pile established by Darcy's law, the speed of rising temperature diminished only with porosity, showing that the impact of porosity on the presence of oxygen is more powerful than its effect on the transfer of heat. A decrease in porosity can provide a small amount of oxygen for oxidation reactions due to increased resistance to flow in the pile, which accordingly inhibit self-heating in a coal pile.

3.3. Effect of the Gas Flow Rate

Figure 8 shows the effect of the gas flow rate on CPT characteristics. It shows that the rate of gas flow has a minor effect; CPT declined by less than 5 degrees by increasing the gas flow rate from 0.1 to 0.4 l/min, and it was attained in under 10 min. Another interesting observation from Figure 8 is that at the lower gas flow rate, the CPT was not reached. In other words, with this simulation, we can obtain the minimum gas flow rate for avoiding the occurrence of CPT.
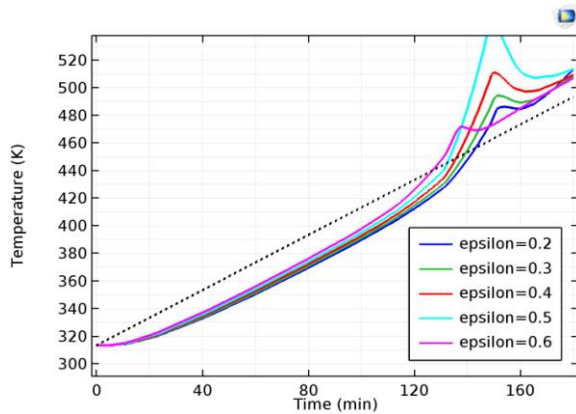
Figure 7 Temperature evolution at 21 vol. % O_2 at an initial temperature of 313.15 K with a gas flow of 0.2 L/min at various porosities
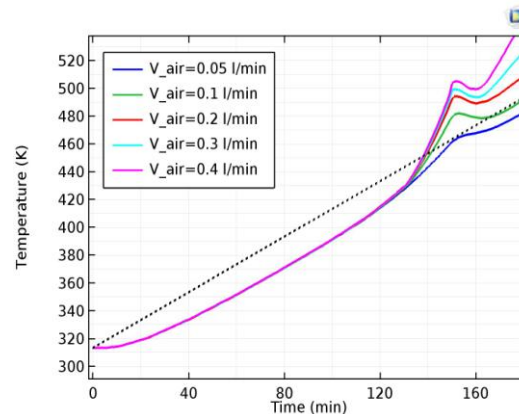
Figure 8 Temperature evolution at 21 vol. % O_2 at an initial temperature of 313.15 K and various gas flow rates

3.4. Effect of Oxygen Concentration

The effect of oxygen concentration on CPT characteristics is summarized in Figure 9. The oxidation reaction rate is strongly influenced by the concentration of oxygen and represents an important consideration, as shown in the Arrhenius equation. The oxygen concentrations highly affect the rate of increasing coal pile temperatures in the reactor. These results indicated that the CPT decreases linearly following the oven temperature with an increase in oxygen concentration from 10 to 40 volume %. The minimum oxygen concentration found at which the CPT did not occur was about 7%.
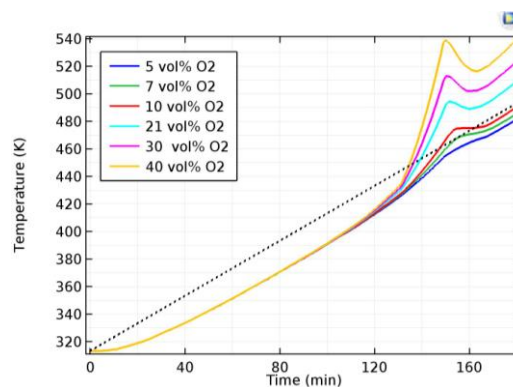

Figure 9 Temperature evolution at an initial temperature of 313.15 K with a gas flow of 0.2 L/min at various oxygen concentrations

4. CONCLUSION

In this research, the CPT phenomenon and parametric studies on the effects of four variables (i.e., initial temperature, concentration of oxygen, the gas flow rate, and porosity) on CPT behavior were investigated using COMSOL Multiphysics. The outcomes of the modeling results agreed with the experimental data, with temperature deviations of about 0.9%. The findings confirmed that porosity and oxygen concentration have major effects on CPT behavior and should be taken into consideration in an attempt to prevent spontaneous combustion.

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