A Percolation-like Phase Transition in Oxygen-limited Combustion

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The combustion process in a mixture of powdered iron and coke is simulated numerically. Complicated combustion patterns appear spontaneously due to the negative feedback effect caused by melted iron preventing the flow of oxygen. By observing the cluster size distribution of regions deprived of oxygen we find a percolation-like phase transition with the control parameter being the amount of coke.

1. Introduction

Because of their importance in practical applications [1], combustion processes have received a lot of attention recently [2], [3], [4]. In the current work we use numerical simulations to study combustion in a mix of powdered iron and coke, a central process in iron manufacture.

Combustion is characterized by irreversible and highly nonlinear chemical reactions. In order to sustain burning the following three conditions are necessary and sufficient:

1. Availability of inflammable material
2. Availability of oxygen
3. High temperature

In situations when at least one of these conditions is fulfilled imperfectly, we can observe the formation of a variety of non-trivial patterns.

So far the applications of statistical physics approaches to combustion problems have been relatively limited. A well-known example is the percolation model of a forest fire [5]. In that case, oxygen is supplied in sufficient quantity everywhere, but trees (inflammable) are distributed randomly and the temperature becomes high only in the neighborhood of the fire front. Another example is the so-called Kuramoto-Sivashinsky equation for nonlinear chemical reactions including combustion [6]. This deterministic equation is known to produce spatio-temporal chaos. Catalysis, like oxidation of CO on a platinum surface, can be also regarded as a kind of combustion in a very wide sense. Ziff-Glari-Barshad's lattice model of CO oxidation demonstrates the appearance of very complicated patterns in oxygen-limited or CO (inflammable)-limited cases [7].

2. The process of iron manufacture

In the process of iron manufacture, the common procedure is to sinter powder or granular iron ore by burning powder coke which is well mixed with the iron ore. The mixture of iron ore and coke is contained in a box with downward air flow, so combustion proceeds from the top to the bottom. Melting of iron ore at high temperature is required to coagulate the powdered iron, but liquid iron prevents the flow of oxygen and tends to extinguish the fire. If we add too much coke in the iron ore, combustion can cease even at the surface because melted iron completely covers the top of the box and chokes the whole oxygen supply. Because of this negative feedback effect the combustion process becomes unstable and the resulting combustion pattern becomes very complicated when the quantity of coke is intermediate. A remarkable experimental finding is that a percolation-like phase transition occurs at a certain amount of coke and the best sintered state is around the critical point [8]. In this paper we numerically simulate the combustion process of the mixture of iron ore and coke as an example of oxygen-limited combustion. The experimental finding is confirmed.

3. The model

We model the combustion process in a hypothetical 2-dimensional system by breaking the combustion chamber into small squares and mapping it on a square lattice. The quantities we introduce are defined on sites of the lattice and are listed as follows:

$C_{i,j}$: volume fraction of coke
 $I_{i,j}$: volume fraction of iron
 $O_{i,j}$: volume fraction of oxygen
 $T_{i,j}$: temperature

Here subscripts i and j denote the coordinates of a site and $C_{i,j} + I_{i,j} + O_{i,j} = 1.0$. For simplicity heat is assumed to be transferred by thermal diffusion, and the temperature field is governed by the usual discretized diffusion equation with source terms determined by the combustion process. We have chosen the value of the diffusion coefficient for temperature (denoted by $V_{i,j}$) as so large that the temperature around burning sites easily exceeds the inflammation point, T_c . To account for the experimentally known fact that the whole combustion process is limited by oxygen supply, the diffusion coefficient takes a smaller value at regions where the airflow is prevented. The temperature is given by

$$T_{i,j}(t+1) = T_{i,j}(t) + V_{i,j}\{T_{i,j+1} + T_{i,j-1}(t) + T_{i-1,j}(t) + T_{i+1,j}(t) - 4T_{i,j}\} + \Delta_{i,j}(t), \quad (1)$$

$$V_{i,j} = \begin{cases} 0.15, & O_{i,j} > 0.1 \\ 0.05, & \text{otherwise,} \end{cases}$$

$$\Delta_{i,j}(t) = \begin{cases} Q \min(C_{i,j}(t), C_0), & \text{if } (i,j) \text{ is burning} \\ 0, & \text{otherwise,} \end{cases} \quad (2)$$

We assign two threshold values, $T_c (=1)$, for the inflammation point and T_m for the iron melting point ($T_c < T_m$). The coke at (i, j) , $C_{i,j} (\neq 0)$, burns if $T_{i,j} > T_c$ and $O_{i,j} = O_0 (=0.1)$. The amount of coke burnt in one time step is $C_0 = 0.02$ and the local temperature is raised by $Q \times C_0$:

$$C_{i,j}(t+1) = \begin{cases} C_{i,j}(t) - \min(C_{i,j}(t), C_0), & \text{if } (i,j) \text{ is burning} \\ C_{i,j}(t), & \text{otherwise.} \end{cases} \quad (3)$$

For sites with $T_{i,j} > T_m$ iron is regarded to be in a liquid phase and to quickly moves downward due to gravity. By this effect $I_{i,j}$ is distributed to the lower neighboring sites, $(i-1, j+1)$, $(i, j+1)$, and $(i+1, j+1)$, until the destination sites become full:

$$I_{i,j}(t+1) = I_{i,j}(t) - \Delta I_{i,j}, \quad (4)$$

$$\Delta I_{i,j} = \begin{cases} \min(I_{i,j}(t), G_{i,j}) & \text{if } T_{i,j} > T_x \\ 0, & \text{otherwise,} \end{cases} \quad (5)$$

$$G_{i,j} = \sum_{k=i-1}^{i+1} D_k(1 - I_{k,j+1}(t)). \quad (6)$$

$$I_{k,j+1}(t+) = I_{k,j+1}(t) + \Delta I_{i,j} \frac{D_k(1 - I_{k,j+1}(t))}{G_{i,j}}. \quad (7)$$

Here $k = i-1, i, i+1$, $D_{i-1} = D_{i+1} = 1 - (\sqrt{2}/2)$, $D_i = \sqrt{2} - 1$.

Sites full of iron choke the oxygen flow. The oxygen flow is also stopped at burning sites, namely the oxygen is considered to be fully consumed by combustion. Therefore, oxygen is not supplied to (i, j) if all of the three upper sites $(i-1, j-1)$, $(i, j-1)$, and $(i+1, j-1)$ are either burning or full of iron.

$$O_{i,j}(t+1) = \begin{cases} 0, & \text{if } (i-1, j) \text{ and } (i-1, j-1) \text{ and } (i-1, j+1) \text{ are burning} \\ \min(O_{i,j}, \max(O_{i-1,j-1}(t), O_{i,j-1}(t), O_{i+1,j-1}(t))), & \text{otherwise.} \end{cases} \quad (8)$$

Here $\min(x_1, x_2, \dots)$ and $\max(x_1, x_2, \dots)$ denote the minimum and the maximum values in the arguments, respectively.

The simulations are started with uniform initial matter distribution corresponding to $C_{i,j} = 0.2$ and $I_{i,j} = 0.3$, while the temperature at the top is given by uniform random numbers, $T_{i,0} \in [0.8, 1.2]$ for $t = 1$ to 5, $T_{i,0} = 0$ when $t > 5$, $T_{i,1} \in [0.7, 1.1]$, $T_{i,2} \in [0.6, 1.0]$ and $T_{i,j} = 0.0$ if $j > 2$ for $t = 1$. Namely, we initially ignite the top of the powder cake with the temperature randomly fluctuating around the inflammation threshold. The boundary condition in the horizontal direction is periodic. In the vertical direction, a fixed boundary condition is applied, $C_{i,0} = C_{i,255} = I_{i,0} = I_{i,255} = T_{i,255} = 0$. Note that coke and iron ore are perfectly mixed at the start and randomness comes only from the ignition condition.

4. Simulation results

The simulations were done by a SUN SPARC workstation with the system size 256×256 . Fig. 1 shows typical patterns formed by melted iron and by regions deprived of oxygen after combustion has taken place. Fig.

1(a) corresponds to combustion with relatively small combustion heat Q . There are many holes in the iron pattern and small clusters of sites deprived of oxygen are distributed nearly uniformly. For a larger Q larger voids and larger blocks of sites deprived of oxygen appear as shown in Fig. 1(c). These figures are reminiscent of cellular automata patterns [9]. In Fig. 1(d), the value of Q is large enough to allow an immediate formation of a layer of melted iron, blocking the combustion process at an early stage.

As mentioned above, the evolution rule is deterministic and the initial distribution of matter is uniform. Therefore, the complicated mass fluctuation, observed at the intermediate values of Q (Fig. 1(b) and Fig. 1(c)), results spontaneously from the small fluctuation in the ignition condition at the upper boundary.

The quantity of primary practical interest, the total mass of melted iron, is plotted versus Q in Fig. 2. Here, averages are taken over 10 realizations. For $Q < 18.0$ we have almost no melting while the coke burns out nearly perfectly. The quantity of melted iron increases suddenly around $Q = 20.0$. Then the behavior becomes somewhat complicated because the increase in combustion heat, on the one hand, leads to the increase of the mass of melted iron in the bulk while, on the other hand, it increases the probability that the melted iron blocks the oxygen supply at the upper layer, suppressing combustion in the bulk, and thus decreasing the overall amount of melted iron (this kind of blocking is clearly seen in Fig. 1(a) and Fig. 1(b)).

For small enough Q , the pattern of melted iron (or of regions deprived of oxygen) is formed by small (comparing to the size of the system) clusters of approximately the same size. As Q approaches 28, blocking of oxygen supply at the upper layer leads to the appearance of large areas deprived of oxygen, neighboring the segments of the upper layer, covered by melted iron. At the same time, the area that is not screened by melted iron in the upper layer displays largely the same pattern of melted iron (regions deprived of oxygen) as in the case of a smaller Q (Fig. 1(a) and Fig. 1(b)).

Fig. 1. Patterns formed by melted iron (above) and by the regions deprived of oxygen (below), formed at different values of Q (the combustion heat). (a) $Q = 24$. (b) $Q = 28$. (c) $Q = 36$ (the phase transition point). (d) $Q = 38$.

As Q is increased still further, close to $Q = 36$, while the blocking effect at the upper layer is retained, the appearance of the active area is drastically changed (Fig. 1(b) and Fig. 1(c)). Now, we can observe a wide distribution of cluster sizes, including clusters with size of the order of the size of the system. So there are two maxima, at $Q = 26$ and at $Q = 36.0$. For $Q > 36.0$ the combustion process is blocked nearly immediately, the overall quantity of melted iron is low and proportional to the linear size of the system.

Now, we would like to concentrate on the behavior of the system near $Q = 36.0$, corresponding to the global maximum of the amount of melted iron. As one can note in Fig. 1(c), this case is characterized by the presence of clusters of melted iron and clusters of sites deprived of oxygen of a size comparable to the size of the system.

To further clarify the behavior around the global maximum, we plot the cumulative cluster size distribution for the regions deprived of oxygen on a log-log scale (Fig. 3). There is a rapid decay at $Q = 22.0$; the size distribution is convex. At $Q = 36.0$ the distribution apparently follows a power law, which is approximately given as $P(>s) \propto s^{-0.97}$, while for $Q = 38.0$ the distribution is concave. By analogy with cluster size distributions in critical systems, we judge that our system shows some kind of critical behavior at $Q_c = 36.0$.

The dynamic phase transition we observe here has some similarity with the static percolation transition [5]. For $Q < Q_c$ we have only small clusters of sites deprived of oxygen. At $Q = Q_c$ the cluster size distribution follows a power law and for $Q > Q_c$ there appears a spanning region deprived of oxygen, corresponding to the infinite cluster in the usual percolation. This result is consistent with the experimental observation of sinter

percentage of melted iron

Fig. 2. Total percentage of melted iron versus the value of parameter Q .

$\text{Log}(N) \{N:\text{size} \Rightarrow \text{SIZE}\}$

Fig. 3. Cumulative size distribution for clusters of sites deprived of oxygen.

cakes using X-ray CT technique scanning [8].

5. Conclusions

We have simulated pattern formation in oxygen-limited combustion of a mixture of iron ore and coke. The choking effect of the melted iron makes the system very unstable. We found a percolation-like dynamic phase transition. The quantity of melted iron is maximal near the critical point, namely, the efficiency of sintering is best around the critical point. Although our simulation was performed on a 2-dimensional lattice, this result is consistent with 3-dimensional experiments.

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