Synchronized smoldering combustion

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Abstract – Synchronized, pulsating temperatures are observed experimentally in smoldering fires. The entire sample volume (1.8 l) participates in the pulsations (pulse period 2–4 h). The synchrony lasts up to 25 h and is followed by a spontaneous transition to either disordered combustion or self-extinguishment. The synchronization is obtained when the fuel bed is cooled to the brink of extinguishment. Calculations for adiabatic conditions, including heat generation from combustion (nonlinear in temperature) and heat storage in sample (linear in temperature), predict diverging sample temperature. Experimentally, heat losses to surroundings (linear in temperature) prevent temperatures to increase without bounds and lead to pulsations.

Introduction. – Synchronized systems have gained significant attention recently, due to the large number of systems that display synchronization and new insights into the mechanisms that lead to and maintain synchrony [1–3]. A synchronized system exhibits the same behavior throughout the system, like soldiers marching in step. Synchronized systems are frequent in nature: the human cardiac rhythm is synchronized with the respiratory system [4], lynx and hare populations are synchronized [5], synchronization is observed between solar activity and gravity [6], as well as in microbiological intracellular communication and in cell movement [7]. Synchronization is relevant for nonlinear optics, physiology and fluid dynamics [8]. Synchronization is also important for understanding and controlling networks, like high-tech industrial networks, large social groups and brain activity [9,10]. Synchronization can manifest itself in a number of ways, globally or locally, but is challenging to obtain experimentally.

We report synchronization during smoldering combustion. Smoldering is a flameless form of combustion, which propagates more slowly than flaming fires and occurs at lower temperatures [11]. Heat is generated through incomplete oxidation at the fuel surface, whereas during flaming oxidation occurs in the gas phase. Smoldering is self-sustained when the heat generated by exothermic oxidation reactions balance heat losses to surroundings. High heat losses can cause self-extinguishment. Smoldering fires also display sudden transitions to flaming fires or explosions [12,13]. Industrially, this is a concern for storage facilities with materials prone to smoldering, like biofuel silos and waste deposits [13,14]. In wildfires and peatland fires, post-flame residual burning may last for days, months or even years [15]. Coal-seam fires cause human relocation, large emissions to the atmosphere and destruction of habitats [16]. The world’s oldest fire has been smoldering through a coal seam deep underground for more than 6000 years [17]. The aerosol and greenhouse-gas emissions from smoldering fires are significant [18]; smoke from smoldering fires worldwide corresponds to more than 15% of man-made greenhouse gas emissions [15,19]. Despite challenges like these, smoldering is a poorly understood combustion phenomenon [11].

Smoldering is a slow, enduring and self-sustained combustion process, disordered in space and time, with long-lived hot regions, called smoldering nests. Smoldering can move steadily through a material, but can also display fingering fronts [20]. In some cases, advancing smoldering
The sample was heated from below a height of 100 mm. The insulated stainless-steel cylinder holding the 1.8 l, 1.25 kg sample. The initial heating regime, where not only heat generated by the process, longer heating period would lead to a different smoldering, while shorter heating was not, as found in preliminary experiments. On the other hand, a sustained smoldering, while shorter heating was not, as found in preliminary experiments. But also excess of external heat drive the evolution. The sample was left undisturbed (with the heater off) until the combustion had ceased and ambient temperature was reached throughout the sample.

Sample temperatures were measured using K-type thermocouples mounted on a small stainless-steel cylinder positioned on a vertical plane near the sample center, with vertical spacing 20 mm and three measurement points at each height (fig. 1 (right)). Heater temperature was measured by a thermocouple placed on top of the heater. A cooling unit consisting of a 4.8 mm outer diameter copper pipe with loop geometry was positioned near the sample center with the lower tip of the loop 10 mm above the heater. The cooling unit and the thermocouple plane were both shifted about 5 mm laterally in opposite directions from the center to avoid direct contact. Water was circulated through the pipe, water temperatures at inlet and outlet, and flow rate were measured. The set-up was positioned on top of a scale. Data was recorded at 5 s intervals.

Results and discussion.

No water cooling. A typical smoldering experiment with no synchronization is shown in fig. 2(a). The sample was heated from below, leading to slowly increasing temperatures, with highest values in the lower part of the sample throughout the external heating. A self-sustained smoldering fire was initiated, indicated by temperature increase above the imposed heater temperature in multiple measurement positions after the heater was switched off at 13 h. Approximately 120 experiments have been carried out with wood pellets in the geometry shown in fig. 1 without the cooling unit. All display disordered combustion with steady or intense consumption of fuel like in fig. 2(a), with no synchronization. Also for experiments with cooling unit in the sample center (see fig. 1) but without water, disordered combustion with no synchronization was observed, as for the experiment in fig. 2(a). There were conductive heat losses through the cooling unit, but without effects on the overall combustion pattern.

Strong cooling, pulsations. With water flow through the cooling unit from the start of the external heating, the expected disordered combustion was not observed immediately after the heater was switched off. Instead, the system cooled down for 2–3 h, before the temperatures started pulsating (fig. 2(b) and (c)). During a pulse, the temperatures increased in a synchronized manner throughout the sample, reaching typical temperature maximums of 300–500 °C, with pulse duration of 2–4 h. After reaching maximum temperatures, the entire fuel bed cooled down and remained so for several hours, before the temperatures in concert again increased towards a new pulse. Here lies the synchrony.

In the experiment displayed in fig. 2(b), there were 9 consecutive synchronized pulses, after which the system displayed a spontaneous transition from synchronization to disordered intense combustion at 42 h. In fig. 2(c), there

Fig. 1: Diagram of experimental set-up (left): the sample was held in a steel cylinder with insulated side walls and heated from below. The sample was cooled by water flowing through a copper pipe near the sample center. Thermocouple positions (right): sample temperatures were measured in a vertical plane near the sample center: horizontal positions left (L), center (C) and right (R), vertical spacing 20 mm. Heater temperature was measured at 0 mm center. All distances are in mm.
Synchronized smoldering combustion

Fig. 2: Temperatures as a function of time. Legend (given in (d)): red is the heater (at 0 mm), the other color codes give the vertical height above the heater (20–100 mm) and the line type gives the horizontal position (L, C, R given in fig. 1). Center temperatures are affected by the nearby cooling unit. Heater-off at 13 h is indicated by the vertical line.

(a) No water cooling, no water in the cooling unit: disordered combustion. (b) and (c): strong cooling, water flow through the cooling unit: respectively 9 and 4 synchronized temperature pulsations, followed by transition to disordered combustion. (d) Strong cooling: two temperature pulsations followed by self-extinguishment.

were 4 pulses before a transition to disordered combustion at 24 h. During the pulsating period, hot regions traveled within the sample (horizontally and vertically) at median traveling speeds of 2.1 mm/min (see the supplemental material for video animations cited in the appendix). This was not observed during disordered combustion, and interestingly, the traveling speed resembles the median of 1.8 mm/min for smoldering fronts in horizontal fuel beds [11]. From the large number of performed experiments, the rare phenomenon of repetitive pulsations (shown in fig. 2(b) and (c)) was observed in 7 out of a total of 14 experiments with strong cooling (pulsation period 17.5 ± 5.5 h). In the remaining 7 experiments, there were one or two pulsations, followed by self-extinguishing, see fig. 2(d) (pulsating period 4.7 ± 2.1 h). No active extinguishing measures were used other than the central cooling unit with water flow. The variation in experimental outcome, despite the same initial conditions reflects the stochastic nature of smoldering.

Runaway pulsations. For the pulsating period, linear regression showed a significant ($p < 0.05$) increase in both maximum temperature and pulse frequency as the system approached disordered combustion, see fig. 2(b),(c). Including all pulsating experiments, the last pulse before disordered combustion had significantly higher temperatures and frequencies (413 ± 44 °C and 0.53 ± 0.2 pulses per hour) compared with the pulsations followed by a new pulse (367 ± 55 °C and 0.34 ± 0.1 pulses per hour). Significance is determined at $p < 0.05$ by a two-tailed Mann-Whitney U test [25]. This indicates an increasing activity in the system as it approaches disordered combustion. The systematic increase also indicates that for a given pulse, there is some degree of predictability of whether the pulse will lead to a disordered situation, or maintain its synchronized combustion. Self-extinguishment could not, however, be predicted based the temperature or frequency of a pulse. The temperatures lie in the same range as the first pulsations of the non-extinguished pulsations (although often in the lower end of the range, as in fig. 2(d)). No increase in frequency could be determined with significance due to too few pulsations before self-extinguishment.

Low-intensity combustion. Comparing pulsations with disordered combustion, the pulsations represent a low-intensity form of combustion: Mass loss rates were an order of magnitude lower for the pulsating period than for the disordered period (typical ranges 25–45 g/h vs. 140–600 g/h). The maximum temperatures were also lower, 379 ± 55 °C vs. 599 ± 33 °C.

Stability criterion. To further elucidate the transition from pulsating to disordered combustion, consider the stability criterion used for a quasi–two-dimensional smoldering front, in terms of the dimensionless quantity $Ar_c = RT_c/E$, where $R$ is the gas constant, $T_c$ is the combustion temperature and $E$ is the activation energy, a material-specific property [21,26]. These fronts oscillate, in the sense that propagation velocity varies, over positive values, in an oscillatory way. The criterion gives a material-specific prediction for transition from oscillating ($Ar_c < 0.03$) to non-oscillating smoldering ($Ar_c > 0.03$), facilitated by a high combustion temperature. For our case, where we have measured the activation energy 91.4 kJ/mol [27], the corresponding $Ar_c = 0.034 ± 0.005$ for
Fig. 3: Heat transfer during the pulsating period (15–24 h) for the experiment shown in fig. 2(c). Net power > 0 denotes heat loss, < 0 heat gain or production. (a) The governing contributions behind the pulsations: heat loss to the cooling unit (blue dashed line), heat production from combustion (red dotted line) and sum of heat losses (black full line). (b) Sample temperatures (see fig. 2 for legend), vertical lines indicate maxima in the average temperatures of each peak, to enable peak comparison.

The pulsating period and $0.054 \pm 0.003$ during disordered combustion. Although dimensionality and geometry are different in our system, it is striking that our pulsation period gives an $Ar_c$ value close to the one for the transition from oscillating to non-oscillating mode. The corresponding $Ar_c$ value for the self-extinguished experiments was $0.032 \pm 0.004$.

**Global pulsations.** The thermocouples located within the sample only covered the plane described in fig. 1, to minimize heat conduction along thermocouples and mounting rack. One may object that the pulsations could have occurred only locally near these measurement positions. There are three observations that support the notion of global pulsations. Firstly, based on mass loss and observed sample height during the pulsating period, calculations show that a significant share ($> 40$ vol%) of the sample has been involved in the combustion. Secondly, the thermocouples are placed in different locations (albeit in a plane) and they are all synchronized. Thirdly, further experiments were made with thermocouples mounted on an additional stainless-steel rack along the perpendicular plane to that shown in fig. 1. Synchronized temperature pulsations were observed also in these experiments, supporting the idea that the pulsations indeed involve the entire sample volume.

Pulsations are caused by external cooling. The only physical difference between the experiments with and without pulsations is the centrally located cooling unit. The cooling effect from this cooling unit (fig. 3(a)) was $\sim 18\%$ of the total heat losses during the pulsating period. Cooling peaked at the maxima in the average temperature in the sample (indicated by the vertical gray lines in fig. 3) and at the heat production maxima (fig. 3(a)). The relatively low cooling power, 5–20 W, was sufficient to established a radial temperature gradient and a centrally located heat sink. The cooling power was in the same range as the 0–60 W heat production from combustion. As the sample temperatures increase, eventually the net heat losses from the sample cancel out the driving force, the heat production, causing the maximum-point turnaround of the pulsations. Other main contributors to the total heat loss were thermal radiation heat loss ($\sim 17\%$), heat losses through the insulated container wall ($\sim 8\%$), heat loss due to the buoyant smoke emission at the top of the sample ($\sim 17\%$) and heat loss through the bottom plate below the sample ($\sim 32\%$). Neither could have caused pulsations, since all set-up components but the cooling unit were also used for the experiments with no pulsations.

**Adiabatic model.** The effect of heat losses on the maximum point turnaround can be illustrated by assuming adiabatic conditions. With no heat losses, the heat production in the sample,

$$q_{\text{prod}} = H_c \, m_s \, A_A \, e^{-\frac{E}{RT_i}},$$

(1)

equals the heat storage in the sample,

$$q_{\text{storage}} = \frac{T_{i+1} - T_i}{t_{i+1} - t_i} \, C_p \, m_s,$$

(2)

where $H_c$ is the heat of combustion (6 kJ/g [11]), $m_s$ the sample mass, $A_A$ the pre-exponential factor, $E$ the activation energy, $R$ the gas constant, $T_i$ the sample temperature at time $t_i$, $C_p$ the specific-heat capacity. Using
Pulsation troughs. Large heat losses compared to heat production could be expected to lead to complete extinguishment. For half of the 14 cases with strong cooling it did, but the remaining 7 displayed repetitive pulsations eventually leading to disordered combustion. To understand this surprising behavior, consider the mechanism causing the following minimum-point temperature turnaround. Different mechanisms act at pulsation peaks and troughs, as indicated by the differences in temperature slopes before and after a peak (curves resemble relaxation oscillations, see e.g. ref. [31]). The temperature slope leading up to a temperature peak follows the near-exponential curve given in fig. 4 from eqs. (1) and (2) while the declining temperature slope is less steep, as is expected for cooling [32]. The minimum-point temperature turnaround is most likely linked to the gradual subsidence and local collapses in the sample during the preceding pulsation peak combustion. A pallet grain consists of compacted wood dust, and will after enduring a significant mass loss become too brittle to retain its structural integrity, and disintegrate. Local collapses can give delivery of unburnt fuel to reaction zones, which can cause temperature turnaround in smoldering fuel beds [33]. Gradual compacting of the sample gives more insulated reaction zones with lower heat losses, which also promote combustion. However, compacting may also promote extinguishment, through lower oxygen supply to reaction zones. Combined with the relatively low temperatures of the pulsation troughs, which give lower reaction rates and reduced air convection, both promoting extinguishment, the system is at a balance point. In half the cases, the outcome is self-extinguishment, the other half results in gradual temperature build-up towards new temperature peaks.

At a balance point. It could have been expected that the heat production to heat loss ratio was significantly lower for the cases resulting in self-extinguishment, but this was not the case. The system is at a balance point, with only small variations and slightly lower ratios observed in the self-extinguished cases. Gradual increase in maximum temperatures and pulse frequencies as the pulsations progress also reflect this delicate balance. The cooling unit removes heat generated by smoldering processes, until the heat losses either overcome the heat production and the system self-extinguishes, or the pulsating combustion continues at the balance point until the combustion processes eventually overcome limitations by oxygen supply and heat losses, resulting in disordered combustion. We therefore propose that the synchronized pulsating behavior (fig. 2(b), (c)) is an intermediate state between the normal disordered smoldering process (fig. 2(a)), and the situation where cooling leads to self-extinguishment (fig. 2(d)). This is supported by experiments at a lower degree of cooling (to be reported elsewhere): only the strongest cooling resulted in self-extinguishment. Pulsations were not observed in any experiments without cooling.

Conclusions. – A new smoldering mode has been reported, with synchronized temperature pulsations throughout the fuel bed (1.8 l, 1.25 kg sample), maintaining synchrony up to 25 h. This was obtained by cooling the fuel bed to the brink of extinguishment. The pulsations have increasing pulse frequency and peak temperature as the system approaches disordered combustion (runaway pulsations). Still, the peak-to-peak evolution is to some degree predictable. The pulsating smoldering combustion is proposed to be an intermediate state between normal, disordered smoldering and self-extinguishment.

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APPENDIX

Animations of the temperature distributions along a vertical plane in the sample center from the start to the end of experiments are given. Figure 5 (right) shows a snapshot from an animation. Thermocouple measurement positions are given as circles, linear interpolation is used between the measurement positions. No extrapolation has been made, thus, the 37.5 mm region between the outmost measurement position and insulated container wall is not displayed, nor is the space below the 20 mm vertical measurement position, see fig. 5 (left).

– Animation of fig. 2(a): disordered smoldering, https://risefr.no/media/supplement-fig2a.gif.
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