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Long-Duration Transmission of Information with Infofuses**

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TOC graphics

**“Infofuse” for long:** “Infofuses” – chemically based systems for non-electronic communication that can transmit alphanumeric information encoded as pulses of light – can now operate for hours without accidental extinction. These characteristics improve their functionality and potential for practical use, and make them more convenient as a test bed for a new approach to information technology that fuses chemistry and information—“infochemistry”.

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This paper describes a new system for non-electronic communication that can transmit alphanumeric information as encoded pulses of light, over intervals of hours. The objective of this research is to design “infofuses” that improved the previously described systems\[1\] in three ways: i) they could transmit information for hours instead of seconds, ii) they could transmit long messages, and iii) they resisted accidental extinction. These characteristics improve the functionality and potential for practical use of infofuses, and make them more convenient to use as a test bed for a new approach to fusing chemistry and information—“infochemistry”.

We consider the elements of a system for manipulating information that comprises seven steps: i) generating the message (either by writing it, or by collecting it from a sensor), ii) encoding the information in a form that a device can transmit, iii) transmitting the information, iv) receiving the information, v) decoding the information, vi) interpreting the decoded information, vii) acting on this information. In this work, we focus on steps ii) – iii). To simplify the problem, we assume that reception and decoding will be accomplished optically and electronically, and that there are no constraints (that we must consider) on the complexity, cost, or performance of the systems that accomplish these functions. We also assume that generating the message involves a separate set of issues which may or may not be primarily chemical.

Systems based on infochemistry combine the storage and transmission of encoded information with four attractive features of chemistry: i) high energy density; ii) autonomous generation of power that can be used for both sensing and for transmission; iii) no requirement for batteries; iv) facile coupling with certain kinds of chemical sensing. We have described two infochemical systems that do not require external electrical power (they use only chemical interactions or reactions) to transmit alphanumeric information. The first system—which we call an “infofuse”—is based on a strip of flammable polymer (nitrocellulose).\[1\][2] In this system,
patterns of spots of thermally emissive salts encode information. The second—which we call a “droplet shutter”—is a microfluidic device that capitalizes on the high stability of operation of a flow-focusing nozzle to generate bubbles and droplets.[3] In this system, windows in an opaque mask encode information. The combination of optically transparent droplets and windows serve as optical shutters. A third system—a frequency-agile microdroplet-based laser—requires electrical power to operate the pump laser, and is intended for different applications.[4]

In our previous design of infofuses, information was encoded as patterns of ions (Li+, Rb+, Cs+) patterned on a strip of nitrocellulose.[1] Ignition of one end of a nitrocellulose strip initiated propagation of a flame-front (at T $\sim$ 1000 °C) at 2 – 3 cm/s. As this moving hot zone reached each spot containing added metal ions, it caused the emission of light at wavelengths characteristic of the thermal emission of the corresponding atomic species. The pattern of emissive salts, ordered in space, therefore, became a sequence of pulses of light at characteristic wavelengths, ordered in time.

We have described infofuses that used three thermal emitters (the perchlorate or nitrate salts of Li, Rb, and Cs, with Na as an internal standard for intensity) to encode and transmit alphanumeric messages. Because each of these three ions can either be present or absent in a particular spot, there are seven unique combinations ($2^3$-1; we have chosen not to use 0,0,0 to avoid ambiguities between a pulse and a space between pulses). Seven unique pulses allow an encoding scheme that assigned alphanumeric characters to combinations of two ($7^2 = 49$) sequential optical pulses.[1,5]

Infofuses fabricated from nitrocellulose have three useful characteristics: i) nitrocellulose burns cleanly: while burning, it produces little to no smoke, and the only significant emissive background from burning nitrocellulose comes from adventitious sodium and potassium in the
film. ii) nitrocellulose is easy to process: it is soluble in organic solvents (e.g. acetone, 2-butanone, and methanol) and forms homogeneous films by casting from solution.\(^6\) iii) the intensity of the isotropically emitted light is high, and the radiative transitions from alkali metal (and a few other) atoms give sharp (FWHM < 2 nm) spectral lines (at 672 nm for Li, 780 and 795 nm for Rb, and 852 and 894 nm for Cs).\(^7\) The radiation from an alkali perchlorate salt (~ 1 \(\mu\)g) on a burning strip of nitrocellulose (~ 1 mm wide) can therefore be detected and correctly characterized from any angle at distances as great as ~ 600 m at night.\(^8\)

The infofuses fabricated according to this design demonstrated the principle of a successful strategy for coupling chemistry with the encoding and transmitting of chemical information, but suffered from a number of weaknesses. Two were of primary concern to us: i) While burning, the flame-front had to remain far (> 1−2 mm) from surfaces, so that the heat transfer from the flame to the surface did not cool and extinguish the flame. When the flame-front of nitrocellulose infofuses with dimensions we used (1 – 3 mm wide, ~ 100 \(\mu\)m thick) came close (< 1−2 mm) to any solid surface, their rate of propagation decreased, and the flame frequently extinguished. This characteristic required that the burning infofuses have a vertical orientation, and be out of contact with solid surfaces; these restrictions limited the practical applicability of these infofuses. ii) The rapid propagation of the flame-front (~3 cm/s) precluded times of transmission longer than ~ 1 minute (An infofuse on which the flame propagated at this rate would require a length of 2.6 km to transmit continuous or repetitive messages for 24 hours).\(^9\)

This paper describes a new experimental platform for infofuses that addresses these weaknesses by using a “dual-speed” arrangement (Figure 1), in which a fuse that burns slowly and continuously (a “SlowFuse”) and that does not transmit information, intermittently ignites
fuses (strips of nitrocellulose) that burn quickly and transmit information (“FastFuses”). This design enables us to repeat a message, or transmit different messages, periodically. The design we use here – in which FastFuses are supported on a thermally insulating support (fiberglass), or separated from the surface by some other methods (e.g., crimping) – also gives greater mechanical and thermal stability to the devices, and allows it to be used in a range of orientations.

**Minimizing Chemical and Thermal Interactions Between NC and a Substrate.**

Protecting nitrocellulose infofuses from extinguishing upon interaction thermally with a substrate is critical to their application in transmission of information. Flat fuses ignited on flat thermally insulating polymers, such as Kapton \((k = 0.12 \text{ W (mK)}^{-1})^{[10]}\) or Teflon \((k = 0.25 \text{ W (mK)}^{-1})^{[11]}\) extinguished after ~ 1 – 2 cm of burning, as did fuses ignited on planar substrates of borosilicate glass \((k = 1.0 \text{ W (mK)}^{-1})^{[11]}\) and on aluminized poly(ethylene terephthalate). The observations that flat fuses extinguished on planar glass, and that they burned reliably on fiberglass (vide infra), are consistent with heat transfer from the flame-front to the substrate as causing, in some part, extinction.

There are three potential mechanisms by which extinction could occur: i) heat transfer to the substrate, ii) quenching of the flame by thermally-generated compounds from the fuse or substrate,\(^{[12]}\) iii) mass transport limitations in diffusion and convection of \(\text{O}_2\) to the fuse. Because thermal (and perhaps chemical) interactions between the flame-front of a burning nitrocellulose infofuse and the substrate on which it rests can cool and extinguish the flame, we developed several strategies for minimizing these interactions physically: i) burning it on a good insulator with a low thermal conductivity and heat capacity (e.g. fiberglass or glass wool), ii) crimping the NC fuse (to separate the flame-front physically from the support), and iii) burning it over a trench.
Fuses burned much more reliably on fiberglass than on other substrates. Out of 70 10-cm long, 1-mm wide NC fuses ignited on fiberglass, 65 burned completely.\[^{13}\]\(^{13}\) On other substrates, less than 40 % of the fuses burned completely. Fiberglass has a low thermal conductivity (\(k = 0.04 \text{ W (mK)}^{-1}\)) and high softening point (\(~720 – 850 \text{ °C}\)), and should generate little volatile organics.\[^{14}\]\(^{14}\) Although fuses burned reliably on fiberglass, the proximity of the flame-front to fiberglass did cause the propagation of the flame to slow from \(~2 – 3 \text{ cm/s} to ~1 – 1.5 \text{ cm/s}\). A flame that burned slowly on fiberglass often gave multiplets from a single spot (possibly a source of error) when short integration time (10 ms) for the detector was used.\[^{15}\]\(^{15}\) To ensure that a single spot generates only a single pulse, we either used long integration time (30 – 40 ms) for a flat fuse or fabricated the fuse so that the NC supporting the deposited metal salts could burn at a rate of 2 – 3 cm/s (at the fabricated region, NC has little interaction with a substrate; so the propagation of the flame did not slow down) (Figure S1).

Fuses folded once along their long axis and resting on the substrate in a tent-like configuration resisted extinction by heat transfer: In a set of ten experiments, all of these “tent” fuses (~ 10 cm long, 2–3 mm wide) burned completely while resting on Kapton, but only 30% (3/10) of the corresponding flat fuses burned completely (see Figure S2). Crimping the fuse in this manner prevented the majority of the flame-front from transferring heat to the substrate; only the edges of a crimped NC film contacted the substrate, whereas the much of the surface of a flat NC film did so. As for the fuses on fiberglass, use of crimped fuses in a tent configuration did cause the propagation of the flame to slow from \(~2 – 3 \text{ cm/s} to ~0.5 – 0.8 \text{ cm/s}\) due to heat transfer to the substrate. We achieved a single pulse from a single spot either using long integration time (50 – 80 ms) or a fuse fabricated so the NC supporting the deposited metal salts had little interaction with substrate (Figures 2 and S3).\[^{15}\]\(^{15}\) A complementary strategy, in which a
A 2-mm wide strip of nitrocellulose rested on a 1 mm-wide rectangular trench between two glass slides also protected the flame from extinction (a 1-mm wide fuse resting on a 0.5 mm-wide trench, however, extinguished quickly).

These approaches (crimping the fuse and burning the fuse on a trench) to protecting NC fuses from extinguishing have two advantages: i) they can work on a variety of substrates: it is not necessary to provide a special substrate on which crimped fuses will burn reliably, and ii) crimping the fuses is a simple process that is straightforward to implement. In controlled situations, where an insulating material such as fiberglass is readily available, burning flat fuses on an appropriate material is also a useful tactic. In our opinion, however, crimped fuses are the most general strategy since they can operate while resting on most solid materials without extinction.

A “Dual-Speed” Configuration Allows Infofuses To Transmit Information Over Long Intervals of Time. In addition to the extinction of the infofuses via heat transfer to a substrate, another practical limitation of nitrocellulose infofuses is that they burn at a rate of several centimeters per second: the length of the strip of nitrocellulose determines the length of time that the infofuse can transmit data continuously, and for long intervals of transmission, is impractically long. To overcome this limitation, we developed a “two-speed” configuration for infofuses (Figure 1), in which patterned, fast-burning strips of nitrocellulose (FastFuses) branched off from a central, slow-burning fuse (SlowFuse).

For our SlowFuse, we used a formulation similar to “slow match”, a type of matchcord used in the early days of firearms: cotton string (diameter ~ 1 mm) soaked in a 6% (w/v) aqueous solution of NaNO₃ for ~ 20 minutes and dried at 125 °C. When ignited, the hot zone of this SlowFuse smoldered with a red-hot ember (Figure S4) instead of with a flame, and
propagated at ~ 1 – 2 meters/hour. The SlowFuse emitted a substantial amount of smoke when it smoldered, and although it did glow with a red-hot ember, it did not produce a significant amount of background optical emission, and was not hot enough to cause the embedded sodium to emit. The SlowFuse did not extinguish when burning on fiberglass, and continued to burn even while held loosely between two pieces of fiberglass or while wrapped in glass wool. The smoldering flame of the central SlowFuse was more resistant to extinction than the rapidly burning flame of the FastFuses. In particular, blowing air on the hot zone of the SlowFuse increased the rate at which it burned, while blowing air on the FastFuse extinguished the flame.\textsuperscript{[12]} This design therefore has the characteristic that the extinguishing of one nitrocellulose FastFuse does not prevent the entire system from continuing to transmit information.

For two-speed configuration of fuses, we glued FastFuses to the SlowFuse by applying a solution (~ 5% w/v) of nitrocellulose in acetone and allowing the solvent to evaporate. The SlowFuse was not always able to ignite the FastFuse directly: the heat from the SlowFuse caused the film of nitrocellulose to combust locally (the nitrocellulose disappeared), but without the creation of a self-propagating flame. Small quantities (< 10 mg) of flammable solids at the junction of the two fuses allowed the SlowFuse to ignite the FastFuses (Figure 1). This “handoff” of ignition worked in 100% of trials when we used any of four additives at the junction: i) sulfur (S\textsubscript{8}) powder, ii) magnesium powder, iii) sodium borohydride (NaBH\textsubscript{4}) powder, and iv) sodium azide (NaN\textsubscript{3}) powder adsorbed on the surface of nitrated tissue paper (flash paper).\textsuperscript{[19],[20]} To illustrate the generality of the two-speed approach, we also demonstrated that a cigarette could act as the SlowFuse, with the combination of NaN\textsubscript{3} and nitrated flash paper at the Slow-Fast junction (Figure S5).
To demonstrate using this two-fuse system to transmit information, we appended seven FastFuses to a SlowFuse using flash paper for ignition. Each FastFuse was encoded with its distance from the beginning of the SlowFuse, i.e., the FastFuse that was 1.5 feet from where the SlowFuse was ignited transmitted the message “1.5 FEET”. For this two-speed fuse on a glass wool, the five-foot long SlowFuse did not extinguish until it burned completely (after 45 minutes). Figure 3 shows the transmitted intensity (at a pulse frequency of 6 ± 1 Hz) of lithium, rubidium, and cesium for the fuses that encoded for “1.5 FEET” (which transmitted at 10.7 minutes) and “4 FEET” (which transmitted at 35.7 minutes).

In conclusion, we have described approaches that are major improvements in a previously described system of “infofuses”. Burning fuses on a thermally insulating substrate, or preventing physical interaction between fuses and a substrate, reduced thermal interaction (heat transfer) between fuses and substrate; the fuses thus resisted accidental extinction. We also achieved long-duration transmission of information by joining fast-burning strips of nitrocellulose containing encoded messages to slow-burning cotton string.

This work is important to materials scientists and engineers interested in research at the interface between information science and chemistry — infochemistry. Improved functionality and potential for practical use of infofuses demonstrated in this study could ultimately contribute to achieve infochemical systems that can sense and process chemical or biochemical inputs from the environment and transmit the results optically over a distance.

**Materials and Methods**

Nitrocellulose (11.8–12.2% N) was obtained from Scientific Polymer Products (Catalog no. 711). All metallic salts were obtained from either Alfa Aesar or Sigma-Aldrich in the highest
available purity. All aqueous solutions were prepared with water purified and deionized by a Millipore system. Fiberglass, Kapton, Teflon, borosilicate glass, and aluminized poly(ethylene terephthalate) substrates were obtained from McMaster Carr.

Sheets of nitrocellulose were prepared by pouring ~ 50 mL of a 5% (w/v) solution of nitrocellulose (in acetone) into a 15 cm × 30 cm polyethylene box, closing the lid, and allowing the solvent to evaporate over a few days at room temperature. The resulting sheets of nitrocellulose were either optically clear or slightly translucent. The sheets were then cut into strips with width of 1 – 12 mm using a desktop rotary paper trimmer.

We patterned strips of nitrocellulose with emissive salts via manual spotting. A solution for each of the combinations of unique emitters (Li, Rb, and Cs) of emissive salts for manual spotting were prepared by dissolving ~ 0.5 % (w/v) of the alkali perchlorates in a stock solution of 0.5 % sodium perchlorate in water. Each spot of emitters (~100 nL) was deposited onto the strip of nitrocellulose with a micropipettor (VWR). After all the desired spots were deposited onto the fuse, it was dried in ambient and in an oven at ~ 50 °C for 30 minutes until the water from the deposition of emitters had evaporated.

For spectroscopic detection, a system of four lenses collected and focused the light emitted from a burning infofuse. Two 1-inch diameter fisheye lenses collected light from the entire area occupied by the infofuse; a focusing lens directed the collected light into a 1 mm-diameter multimode optical fiber (Ocean Optics) that was equipped with a collimating lens. Light from this fiber was coupled to a HR2000+ high resolution CCD spectrometer (Ocean Optics), which was connected to a PC with a USB cable and controlled by software (SpectraSuite) supplied by Ocean Optics. Most emission spectra from infofuses were collected
with an integration time of 10 ms (unless otherwise noted), at a rate of 100 spectra/s. The distance between the detector and the burning infofuse was typically 1 m.
Figure 1. a) Schematic diagram of a slow-burning, nitrate-soaked cotton string (SlowFuse), with appended fast-burning nitrocellulose fuses (FastFuses) that transmit information as optical pulses of light characteristic of thermally emissive alkali metals. b) Schematic diagram of a crimped fuse (tent configuration) with little interaction with a substrate.
Figure 2

(a) 5 mm Integration time = 50 ms

(b) 8 mm Integration time = 10 ms

(c) 3 mm Integration time = 10 ms
Figure 2. Two strategies to generate a single pulse from a single spot for crimped fuses (tent configuration) on glass substrate. a) Using long integration time (50 – 80 ms) for collecting data: schematic diagram of a crimped fuse (top), and transmitted light detected from a crimped fuse (bottom). b), c) Fabricating fuses so that the NC supporting the metal salts could have little interaction with a substrate: schematic diagram of a fabricate fuse (top), and transmitted light detected from a fabricated fuse (bottom); Inset images show blowup photographs of fabricated fuses. We colored the fuse blue (part that sits on glass substrate) and red (part where metal salts are deposited) with permanent marker for easy visualization. In all schematic diagrams of fuses, red dots indicate deposited metal salts. In the encoding scheme used here, two consecutive optical pulses represent one alphanumeric character.
Figure 3. Transmitted light detected from a two-speed infofuse when a) a FastFuse ignited after 1.5 feet of the SlowFuse had burned (10.6 minutes), and b) a FastFuse ignited after 4 feet of the SlowFuse had burned (35.7 minutes). In the encoding scheme used here, two consecutive optical pulses represent one alphanumeric character. The fuse burned on a glass wool and was ignited once at $t = 0$ seconds.
References


[5] For detailed encoding scheme, see Supporting Information of ref. 1.


[9] Other weaknesses on which we have not yet focused include low thermodynamic efficiency (For details, see Supporting Information) of coupling the free energy of combustion of nitrocellulose to the emission of photons at frequencies characteristic of atomic emission for the metal ions and sensitivity of the system to the environment (temperature, water content).


[12] Nitrocellulose contains insufficient oxygen to oxidize completely. The oxygen deficit is increased when the solvent is not completely removed from the nitrocellulose. Products of combustion of nitrocellulose such as CO, CO₂, and NOₓ could quench the flame. J. B. Bernadou,
Fuses burned reliably on glass wool, too. Since fuses have little physical interaction with glass wool due to its texture, propagation of the flame maintains the burning rate of ~2 – 3 cm/s on glass wool.

After depositing/drying aqueous solution of metal salt on nitrocellulose infusible, the resulting spot size is ~ 500 μm. For the fuses held vertically (burning rate : ~ 3 cm/s), it takes ~17 ms to burn each spot; so we can achieve a single pulse from a single spot using integration time of 10 ms for the detection. On the other hand, it takes ~50 ms to burn each spot from flat fuses on fiberglass (burning rate : ~1 cm/s); so we can achieve a single pulse by increasing the integration time from 10 ms to 30 – 40 ms.

Fuses crimped into a “tent” configuration must obviously be in the orientation that minimizes contact between the fuse and substrate.

Using dual-speed arrangement of FastFuses with SlowFuses on a 8.5”×11” size fiberglass, we can transmit ~800 characters (with the pulse rate of 10 Hz) of messages for ~ 1 h with efficiency of ~ 0.01 % (for the calculation of efficiency, please see Supporting Information).
2-butyne-1,4-diol, 1,5,9-cyclododecatriene, and other oxidants (nitromethane, hydrogen peroxide, di-tert-butylperoxide, sodium perborate, and sodium pyrophosphate).

Physical methods for “handing-off” ignition from the SlowFuse to the FastFuse were not successful; we tried four methods: i) wrapping NC around the SlowFuse up to three times, ii) resting the FastFuse on the SlowFuse in a collinear arrangement, iii) permeating the SlowFuse with the FastFuse, and iv) wrapping aluminum foil or aluminized mylar around the junction. [20]

The formulations (Nitrocellulose + NaN₃) combine highly flammable materials, strong reducing agents, and oxidizing agents. Although we have not observed any unexpected reactions, care must be exercised in handling such systems. We do not mix the oxidants and reductants directly in solution, and use only several milligrams at a time.
Supporting Information

Long-Duration Transmission of Information with Infofuses

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**Thermodynamic Efficiency of Infofuse**

To calculate the efficiency (total transmitted energy from the burning metal salts/total energy of combustion of the nitrocellulose) of the infofuse, we compared the intensity of emission of a burning infofuse with a green light-emitting diode (LED) of known luminous intensity. At a given distance and at an optimized angle, we measured the integrated intensities of burning fuses and a green LED with known candela output. Then, using the cone angle of the LED (estimated by shining the LED on a piece of paper with trigonometry), the isotropic intensity of the LED has been determined. Comparing the intensity of burning fuses and the LED, we estimated the maximal power of each pulse of an infofuse to be ~ 1 mW.

Heat of combustion of nitrocellulose: $^{20}$ 4.2 kJ/g = 4200 mJ/mg

Density of nitrocellulose: 1.66 g/cm$^3$

For a fuse with width of 1 mm and thickness of 100 µm, mass of a fuse per pulse is (pulse rate of 10 Hz and burning rate of 2.5 cm/s): $(100 \, \mu\text{m}) \times (1 \, \text{mm}) \times (2.5 \, \text{mm}) \times (1.66 \, \text{g/cm}^3) \approx 0.3 \, \text{mg}

Efficiency = Total transmitted energy from the burning metal salts / Total energy of combustion of the nitrocellulose

$$= \frac{1 \, \text{mJ/s} \times 0.1 \, \text{s/pulse} \times (1 \, \text{pulse} / 0.3 \, \text{mg})}{4200 \, \text{mJ/mg} \times 100} \approx 0.01 \%$$
Figure S1. Two strategies to generate a single pulse from a single spot for flat fuses on fiberglass.

a) Using long integration time (30 – 40 ms) for collecting data: schematic diagram of a flat fuse (top), and transmitted light detected from a flat fuse on fiberglass (bottom). b) Fabricating fuses so that the NC supporting the metal salts could have little interaction with the substrate: schematic diagram of a fabricated fuse (top), and transmitted light detected from a fabricated fuse (bottom); Inset image shows a blowup photograph of a fabricated fuse. We colored the fuse blue (part that sits on fiberglass) and red (part where metal salts are deposited) with permanent marker for easy visualization. In all schematic diagrams of fuses, red dots indicate deposited metal salts. In the encoding scheme used here, two consecutive optical pulses represent one alphanumeric character.
Figure S2. Photographs of 3 mm-wide strips of nitrocellulose burning on a film of Kapton. The folded strip (folded once along its long axis) burned completely, while the flat strip extinguished (by heat transfer from the hot zone to the Kapton substrate).
Figure S3. Photographs of fabricated fuses: a) a fabricated fuse in Figure 2b, b) a fabricated fuse in Figure 2c.
Figure S4. Pictures of a burning two-speed infusible resting on fiberglass: a) as the propagating hot ember of the SlowFuse (circled) approached flash paper (nitrated tissue paper), b) as the flash paper burned, c and d) as the FastFuse burned to completion and the burning ember of the SlowFuse continued to propagate (circled). We colored the SlowFuse red and FastFuses blue with permanent marker for easy visualization. The widths of the SlowFuse and FastFuses are ~1 mm, and the lengths of the FastFuses are ~7.5 cm.
**Figure S5.** Using a cigarette as the SlowFuse in a two-speed infofuse. The hot zone of the cigarette ignited the flash paper/NaN\(_3\) combination (d) (flash paper was wrapped around the cigarette with FastFuse using NaN\(_3\) powder and acetone solution of nitrocellulose as glue), which in turn ignited a nitrocellulose FastFuse. The cigarette continued to burn after igniting the FastFuse (the burning rate of cigarette was 0.7 – 0.8 cm/min).