SUPPORTING INFORMATION FOR:

Storage of Information in Mixtures of Fluorescent Molecules

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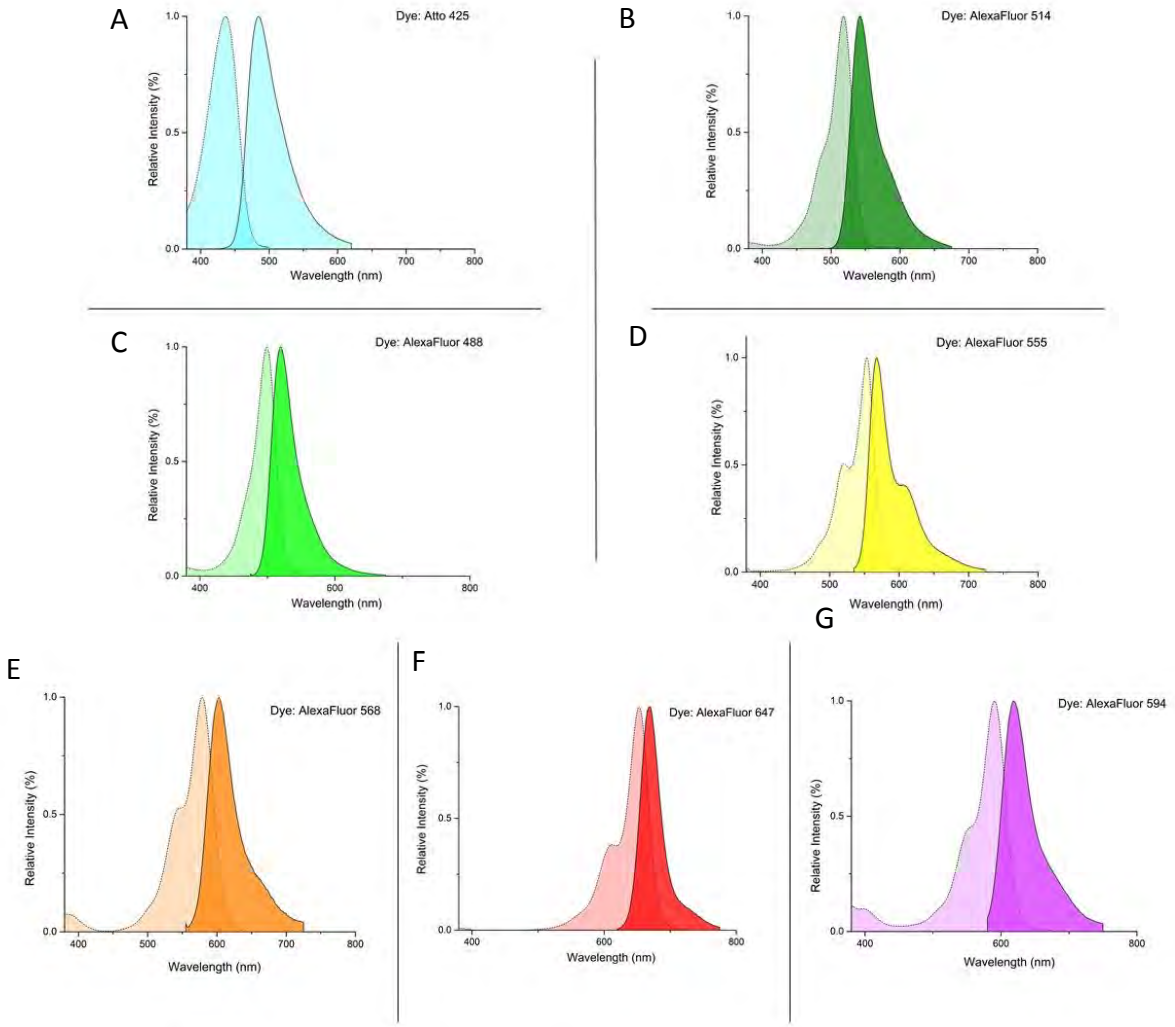

Figure S1. Absorption and emission spectra for (A) dye 425, (B) dye 488, (C) dye 514, (D) dye 555, (E) dye 568, (F) dye 594 and (G) dye 647. Within each plot, the trace on the left corresponds to the absorption spectrum while the trace on the right corresponds to the emission spectrum.

Dye	Concentration
Atto 425	40 µM
AlexaFluor 488	32 µM
AlexaFluor 514	28 µM
AlexaFluor 555	28 µM
AlexaFluor 568	34 µM
AlexaFluor 594	24 µM
AlexaFluor 647	18 µM

Table S1. List of concentrations of the dyes used in this study. All the dyes were purchased as their *N*-hydroxysuccinimide esters.

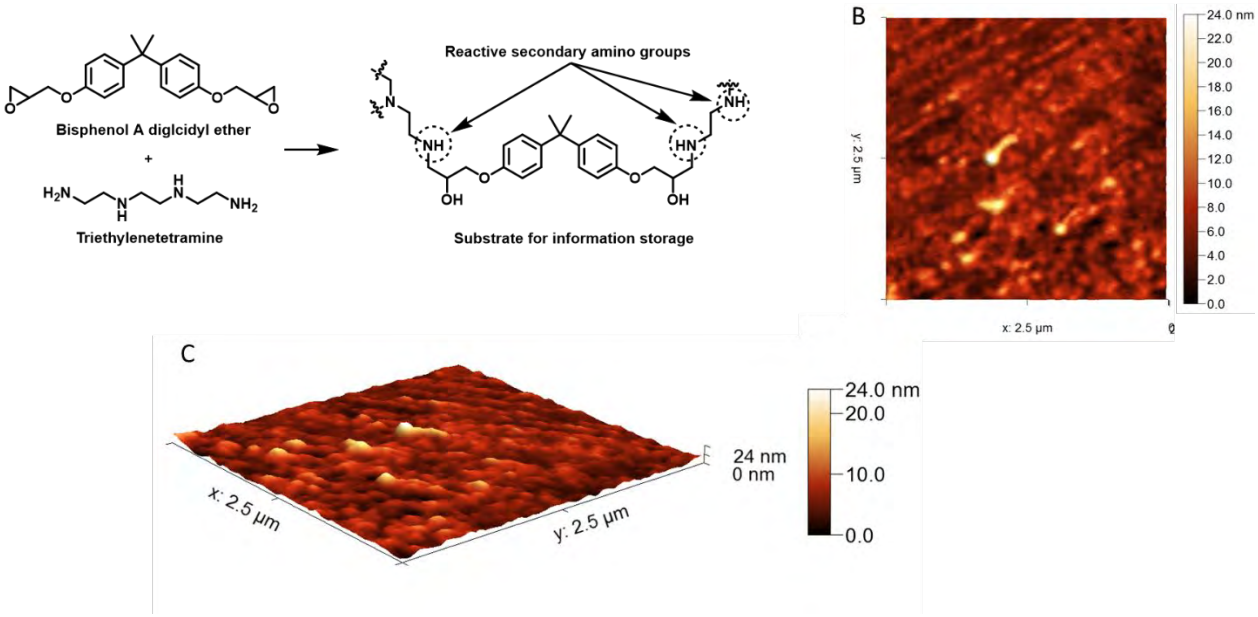

Figure S2. Epoxy substrate. (A) Reaction scheme for fabrication of the epoxy substrate. An excess of the amine (1.2 equivalent with respect to epoxy groups) is used. This composition leads to free secondary amino groups inside the polymer. These secondary amino groups can link the fluorophore covalently inside the polymer by reaction of the active ester of the fluorescent dye molecule with the amino group. (B) Atomic force microscopy (AFM) phase image of the substrate (C) 3D AFM phase image of the epoxy substrate shows the flat topology of the substrate, with an average roughness value of 5.80 nm.

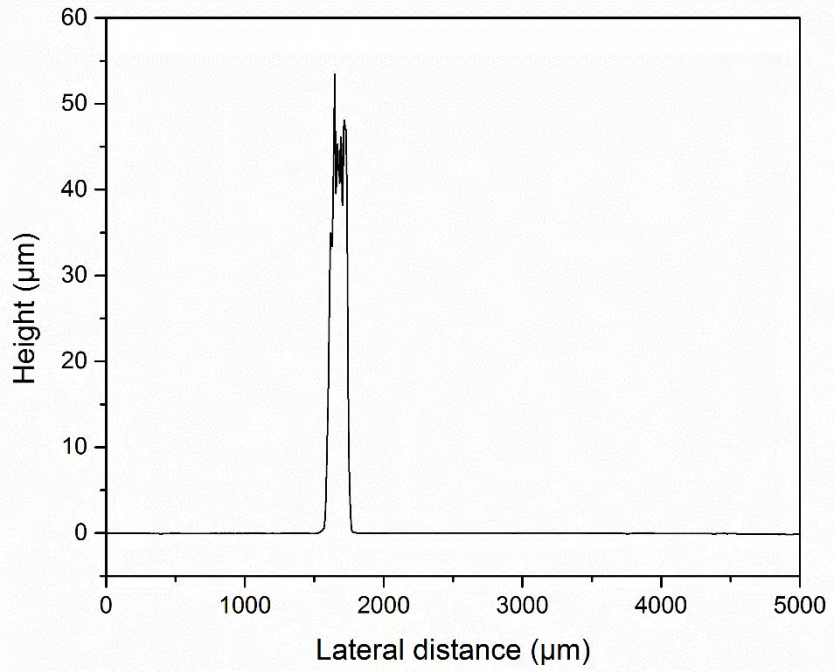

Figure S3. Profilometer data shows that the epoxy substrate is approximately 50 μm in thickness. A sample consisting of an epoxy film on a glass slide was sliced with a razor blade to introduce a cut into the film, sonicated in isopropyl alcohol for 10 minutes, and then dried under nitrogen. Profilometry was performed using a Bruker DektakXT profilometer equipped with a 5-μm radius diamond tip using 3 mg of applied force.

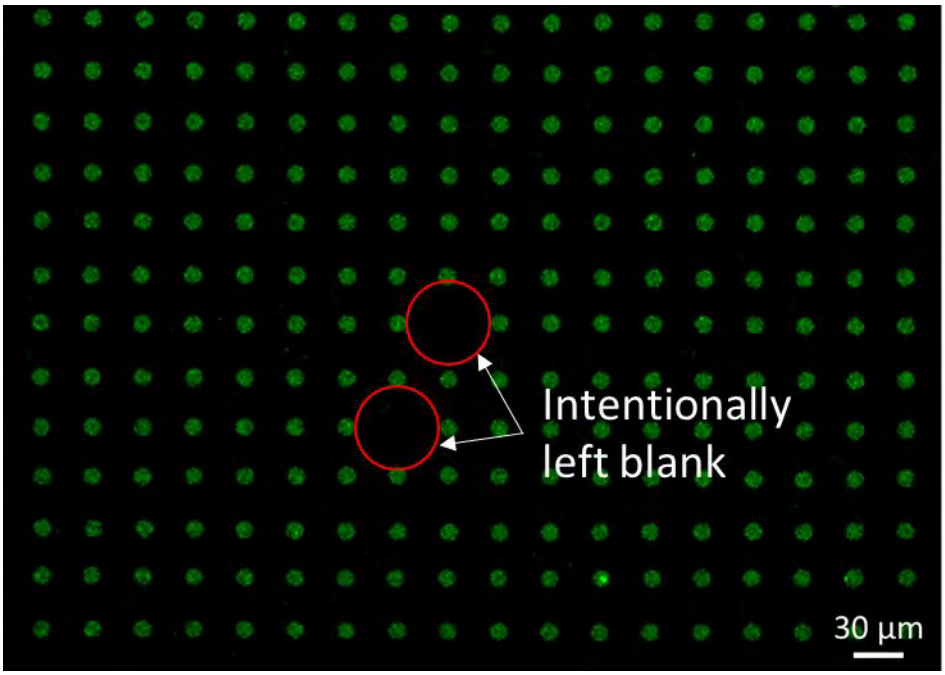

Figure S4. An example of an array of spots inkjet printed with a fluorescent dye – AlexaFluor 488, printed with the Fujifilm Dimatix 2831 printer. The areas in the centre of the array were intentionally left blank to evaluate the accuracy of printing of one dye with our parameters (1 pL drop size, 16V firing voltage).

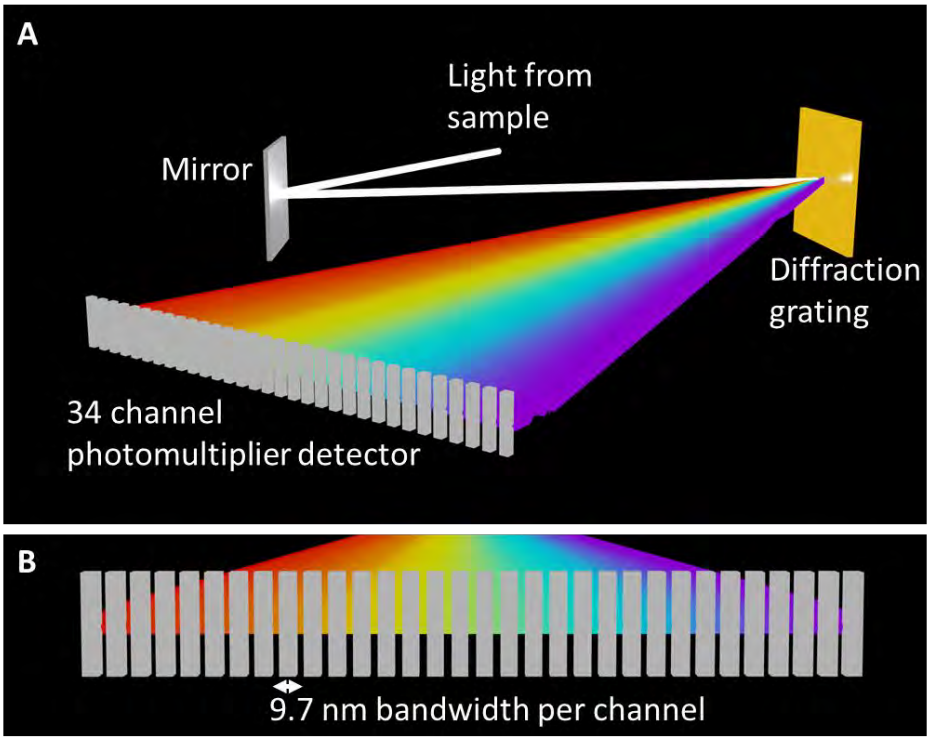

Figure S5: Reading information with a multichannel fluorescence detector. (A) Our "reading" technique uses a spectral imaging setup wherein the fluorescent light beam passes through a pinhole and is reflected on a diffraction grating. The spectrally resolved light is projected onto a 34-channel photomultiplier detector. (B) The wavelength of emitted light is determined by the position of the detection channel receiving the light. Each individual channel covers a bandwidth of 9.7 nm in the visible spectrum.

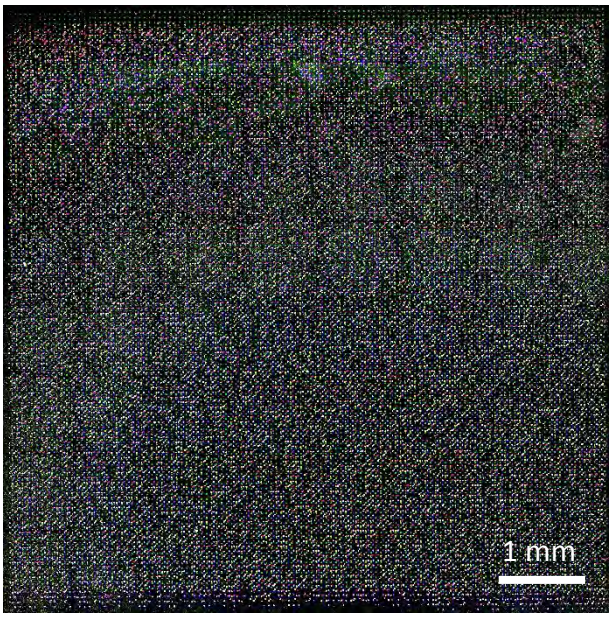

Figure S6. Fluorescence microscope image of the first section of Faraday's "Experimental researches in electricity" on a 50μm epoxy polymer film written using the encoding scheme shown in Figure 3. The image was recorded with excitation using four lasers simultaneously (405nm, 488nm, 561nm, 633nm).

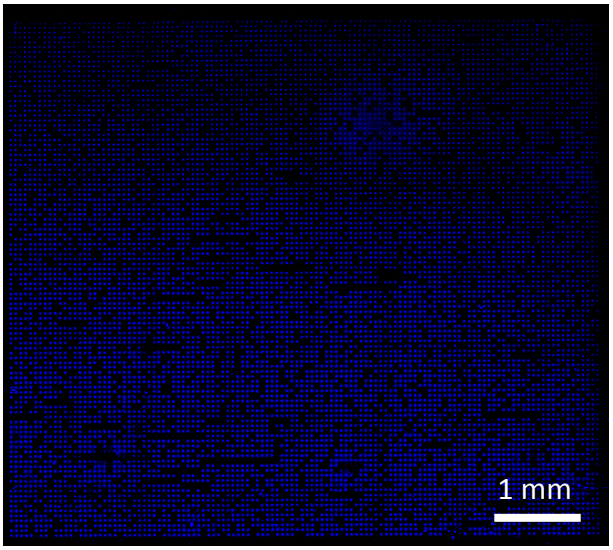

Figure S7. Image of the pattern of the fluorescent dye 425 obtained by spectral unmixing of the original image (**Figure S6**).

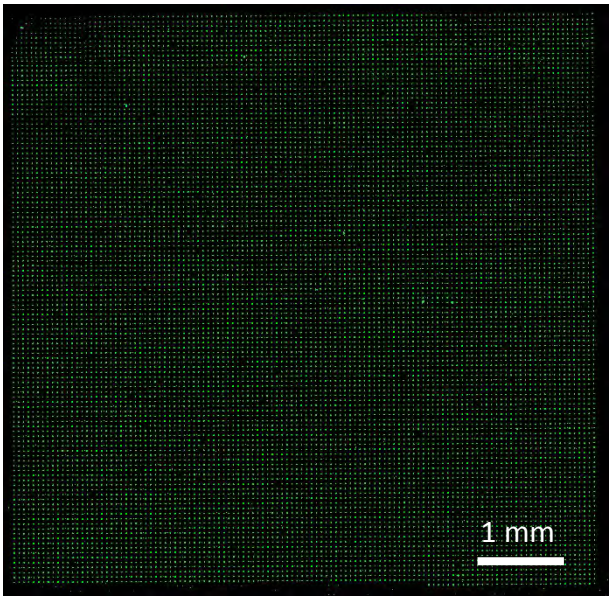

Figure S8. Image of the pattern of the fluorescent dye 488 obtained by spectral unmixing of the original image (**Figure S6**).

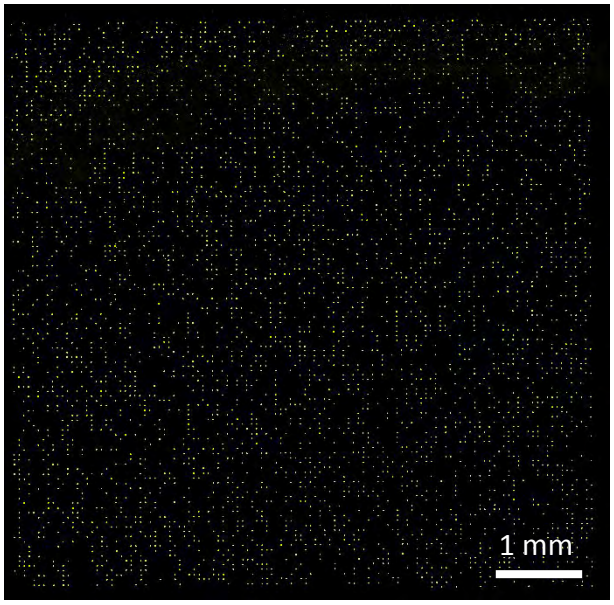

Figure S9. Image of the pattern of the fluorescent dye 514 obtained by spectral unmixing of the original image (**Figure S6**).

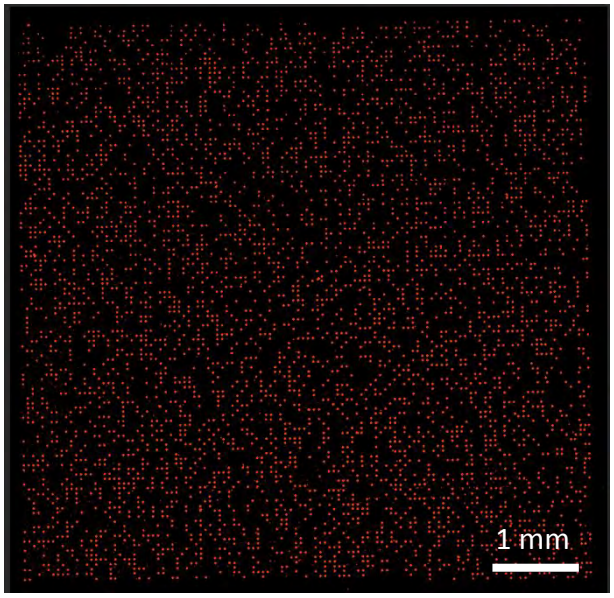

Figure S10. Image of the pattern of the fluorescent dye 555 obtained by spectral unmixing of the original image (**Figure S6**).

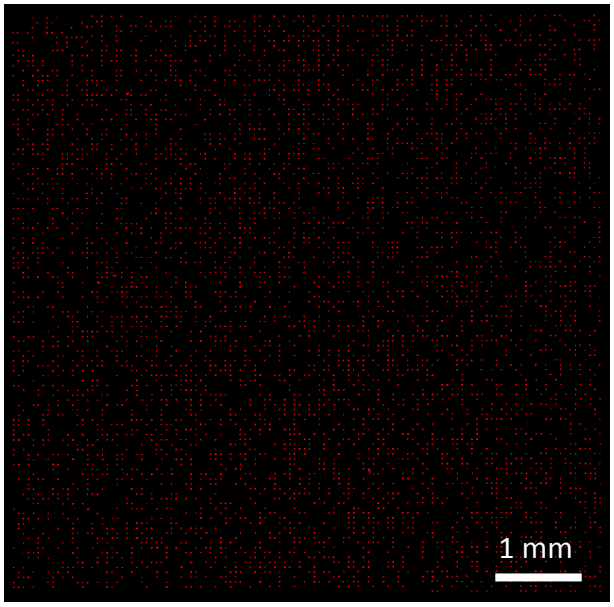

Figure S11. Image of the pattern of the fluorescent dye 594 obtained by spectral unmixing of the original image (**Figure S6**).

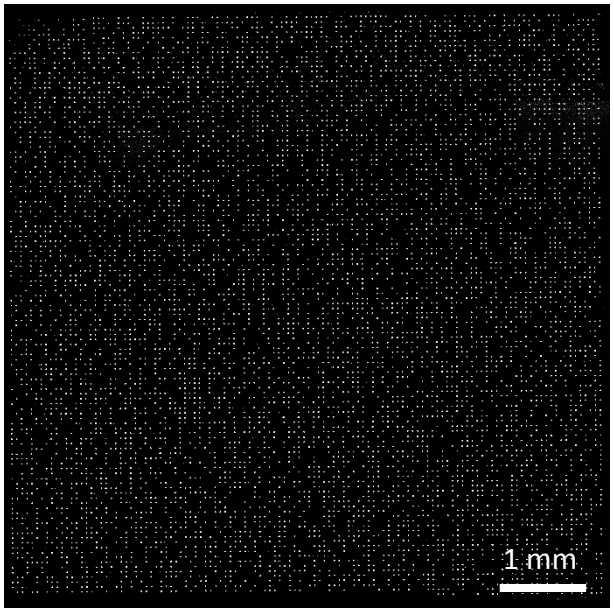

Figure S12. Image of the pattern of the fluorescent dye 647 obtained by spectral unmixing of the original image (**Figure S6**).

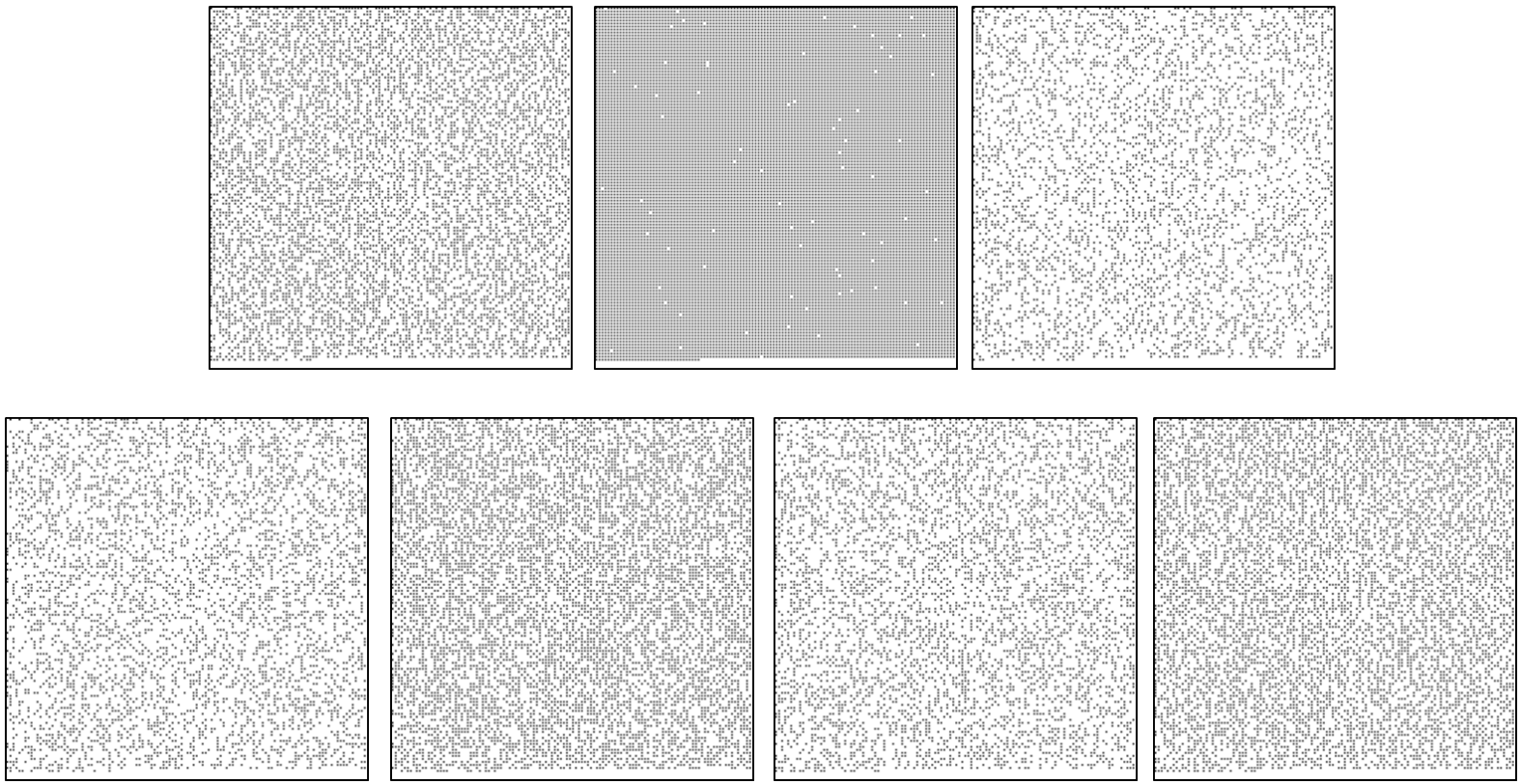

Figure S13. Computer-generated printing patterns for dyes 1-7 for Faraday's paper on electricity. These patterns were printed with the respective fluorescent dyes.

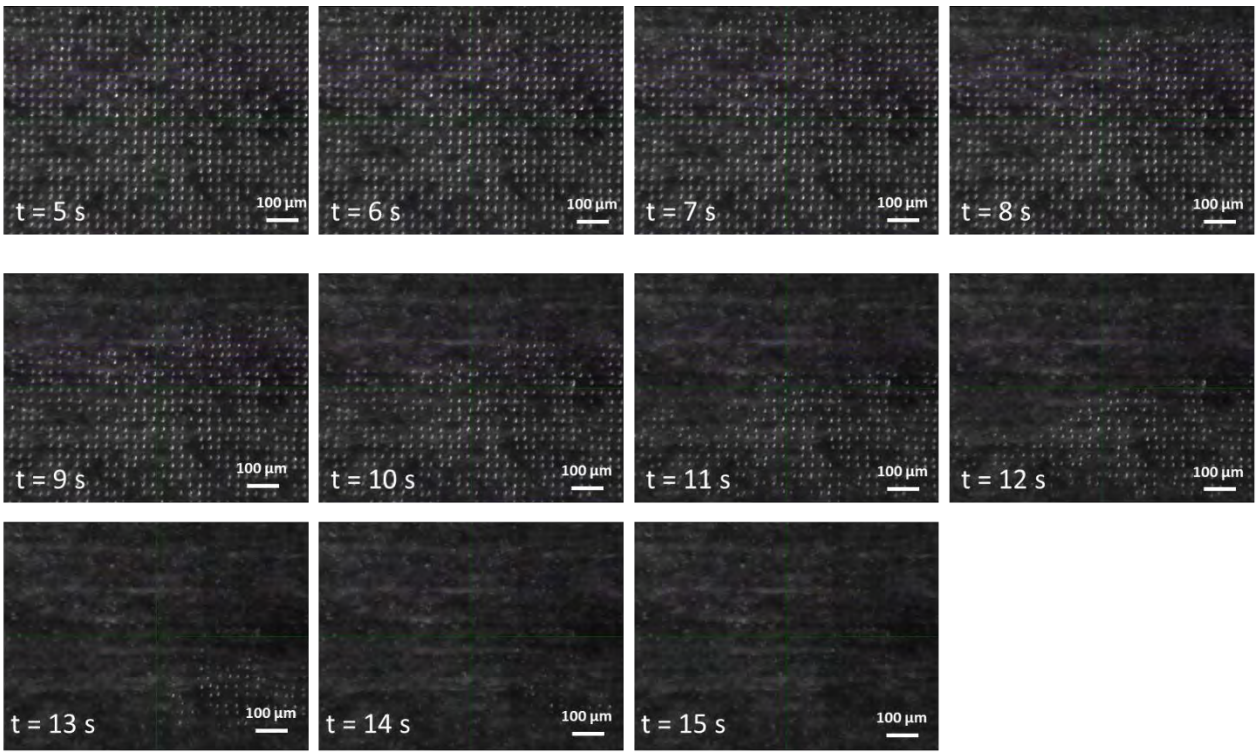

Figure S14. The printed droplets are absorbed inside the epoxy substrate within 15 seconds. Dimethylsulfoxide (DMSO) is used as the solvent to print the fluorescent dye molecules. Images are obtained using the on-board bright field camera inside the Fujifilm DMP-2831 ink-jet printer.

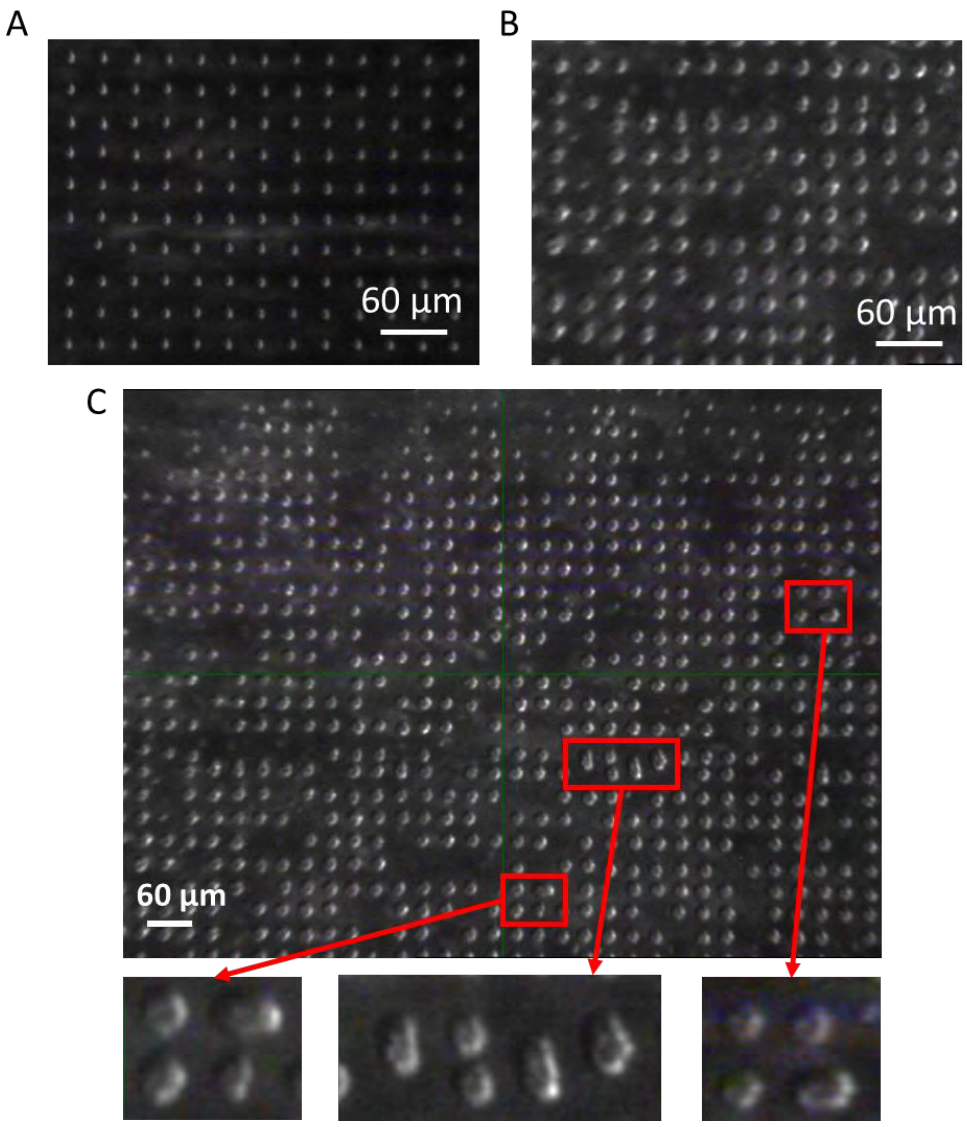

Figure S15. The inkjet printer used in this study supports two droplet volume cartridges – 1 pL and 10 pL drop volumes. Examples of printing of the same dye with different droplet volumes are shown in (A) with 1 pL droplet volume, and (B) with 10 pL droplet volume. The work reported in the manuscript was carried out using the 1pL drop volume cartridges. On printing with larger drop volumes (10 pL), we observed more errors in the printed patterns. For example, figure C shows some of the possible errors that could be observed through the on-board camera inside the ink-jet printer immediately after printing with 10 pL droplets.

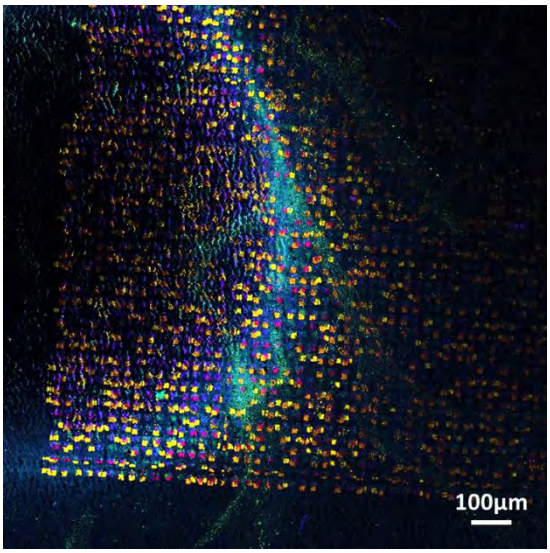

Figure S16. An image of the printed patterns on the substrate when the substrate is not flat. The focused regions look brighter than the non-focused regions. Information in the darker regions is lost. Hence, it is necessary to have a flat substrate to capture all information within one image.

Figure S17. A decoded image of Michael Faraday with 0.4% errors in printing. As the image uses JPEG compression, any raw bit error causes several errors in the final image.

Section S18: Text from the first section of Faraday's "Experimental researches in electricity", Phil. Trans. R. Soc. Lond. 1832, 122, 125-162, that was encoded, written in 7 fluorescent dyes and decoded:

1. The power which electricity of tension possesses of causing an opposite electrical state in its vicinity has been expressed by the general term Induction; which, as it has been received into scientific language, may also, with propriety, be used in the same general sense to express the power which electrical currents may possess of inducing any particular state upon matter in their immediate neighbourhood, otherwise indifferent. It is with this meaning that I purpose using it in the present paper. 2. Certain effects of the induction of electrical currents have already been recognised and described: as those of magnetization; Ampere's experiments of bringing a copper disc near to a flat spiral; his repetition with electro-magnets of Arago's extraordinary experiments, and perhaps a few others. Still it appeared unlikely that these could be all the effects which induction by currents could produce; especially as, upon dispensing with iron, almost the whole of them disappear, whilst yet an infinity of bodies, exhibiting definite phenomena of induction with electricity of tension, still remain to be acted upon by the induction of electricity in motion. 3. Further: Whether Ampere's beautiful theory were adopted, or any other, or whatever reservation were mentally made, still it appeared very extraordinary, that as every electric current was accompanied by a corresponding intensity of magnetic action at right angles to the current, good conductors of electricity, when placed within the sphere of this action, should not have any current induced through them, or some sensible effect produced equivalent in force to such a current. 4. These considerations, with their consequence, the hope of obtaining electricity from ordinary magnetism, have stimulated me at various times to investigate experimentally the inductive effect of electric currents. I lately arrived at positive results; and not only had my hopes fulfilled, but obtained a key which appeared to me to open out a full explanation of Arago's magnetic phenomena, and also to discover a new state, which may probably have great influence in some of the most important effects of electric currents. 5. These results I purpose describing, not as they were obtained, but in such a manner as to give the most concise view of the whole. 6. About twenty-six feet of copper wire one twentieth of an inch in diameter were wound round a cylinder of wood as a helix, the different spires of which were prevented from touching by a thin interposed twine. This helix was covered with calico, and then a second wire applied in the same manner. In this way twelve helices were superposed, each containing an average length of wire of twenty-seven feet, and all in the same direction. The first, third, fifth, seventh, ninth, and eleventh of these helices were connected at their extremities end to end, so as to form one helix; the others were connected in a similar manner; and thus two principal helices were produced, closely interposed, having the same direction, not touching anywhere, and each containing one hundred and fifty-five feet in length of wire. 7. One of these helices was connected with a galvanometer, the other with a voltaic battery of ten pairs of plates four inches square, with double coppers and well charged; yet not the slightest sensible reflection of the galvanometer-needle could be observed. 8. A similar compound helix, consisting of six lengths of copper and six of soft iron wire, was constructed. The resulting iron helix contained two hundred and fourteen feet of wire, the resulting copper helix two hundred and eight feet; but whether the current from the trough was passed through the copper or the iron helix, no effect upon the other could be perceived at the galvanometer. 9. In these and many similar experiments no difference in action of any kind appeared between iron and other metals. 10. Two hundred and three feet of copper wire in one length were coiled round a large block of wood; other two hundred and three feet of similar wire were interposed as a spiral between the turns of the first coil, and metallic contact everywhere prevented by twine. One of these helices was connected with a galvanometer, and the other with a battery of one hundred pairs of plates four inches square, with double coppers, and well charged. When the contact was made, there was a sudden and very slight effect at the galvanometer, and there was also a similar slight effect when the contact with the battery was broken. But whilst the voltaic current was continuing to pass through the one helix, no galvanometrical appearances nor any effect

like induction upon the other helix could be perceived, although the active power of the battery was proved to be great, by its heating the whole of its own helix, and by the brilliancy of the discharge when made through charcoal. 11. Repetition of the experiments with a battery of one hundred and twenty pairs of plates produced no other effects; but it was ascertained, both at this and the former time, that the slight deflection of the needle occurring at the moment of completing the connexion, was always in one direction, and that the equally slight deflection produced when the contact was broken, was in the other direction; and also, that these effects occurred when the first helices were used (6. 8.). 12. The results which I had by this time obtained with magnets led me to believe that the battery current through one wire, did, in reality, induce a similar current through the other wire, but that it continued for an instant only, and partook more of the nature of the electrical wave passed through from the shock of a common Leyden jar than of the current from a voltaic battery, and therefore might magnetise a steel needle, although it scarcely affected the galvanometer. 13. This expectation was confirmed; for on substituting a small hollow helix, formed round a glass tube, for the galvanometer, introducing a steel needle, making contact as before between the battery and the inducing wire (7. 10.), and then removing the needle before the battery contact was broken, it was found magnetised. 14. When the battery contact was first made, then an unmagnetised needle introduced into the small indicating helix (13.), and lastly the battery contact broken, the needle was found magnetised to an equal degree apparently as before; but the poles were of the contrary kind. 15. The same effects took place on using the large compound helices first described (6. 8.). 16. When the unmagnetised needle was put into the indicating helix, before contact of the inducing wire with the battery, and remained there until the contact was broken, it exhibited little or no magnetism; the first effect having been nearly neutralised by the second (13. 14.). The force of the induced current upon making contact was found always to exceed that of the induced current at breaking of contact; and if therefore the contact was made and broken many times in succession, whilst the needle remained in the indicating helix, it at last came out not unmagnetised, but a needle magnetised as if the induced current upon making contact had acted alone on it. This effect may be due to the accumulation (as it is called) at the poles of the unconnected pile, rendering the current upon first making contact more powerful than what it is afterwards, at the moment of breaking contact. 17. If the circuit between the helix or wire under induction and the galvanometer or indicating spiral was not rendered complete before the connexion between the battery and the inducing wire was completed or broken, then no effects were perceived at the galvanometer. Thus, if the battery communications were first made, and then the wire under induction connected with the indicating helix, no magnetising power was there exhibited. But still retaining the latter communications, when those with the battery were broken, a magnet was formed in the helix, but of the second kind (14.), i.e. with poles indicating a current in the same direction to that belonging to the battery current, or to that always induced by that current at its cessation. 18. In the preceding experiments the wires were placed near to each other, and the contact of the inducing one with the buttery made when the inductive effect was required; but as the particular action might be supposed to be exerted only at the moments of making and breaking contact, the induction was produced in another way. Several feet of copper wire were stretched in wide zigzag forms, representing the letter W, on one surface of a broad board; a second wire was stretched in precisely similar forms on a second board, so that when brought near the first, the wires should everywhere touch, except that a sheet of thick paper was interposed. One of these wires was connected with the galvanometer, and the other with a voltaic battery. The first wire was then moved towards the second, and as it approached, the needle was deflected. Being then removed, the needle was deflected in the opposite direction. By first making the wires approach and then recede, simultaneously with the vibrations of the needle, the latter soon became very extensive; but when the wires ceased to move from or towards each other, the galvanometer-needle soon came to its usual position. 19. As the wires approximated, the induced current was in the contrary direction to the inducing current. As the wires receded, the induced current was in the same direction as the inducing current. When the wires remained stationary, there was no induced current (54.). 20. When a small voltaic arrangement was introduced into the circuit between the galvanometer (10.) and its helix or wire, so as to cause a permanent deflection of 30 degree or 40 degree, and then the battery of one hundred pairs of plates connected with the inducing wire, there was an instantaneous action as before (11.); but the galvanometer-needle immediately resumed and retained its place unaltered, notwithstanding the continued contact of the inducing wire with the trough: such was the case in whichever way the contacts were made (33.). 21. Hence it would appear that collateral currents,

either in the same or in opposite directions, exert no permanent inducing power on each other, affecting their quantity or tension. 22. I could obtain no evidence by the tongue, by spark, or by heating fine wire or charcoal, of the electricity passing through the wire under induction; neither could I obtain any chemical effects, though the contacts with metallic and other solutions were made and broken alternately with those of the battery, so that the second effect of induction should not oppose or neutralise the first (13. 16.). 23. This deficiency of effect is not because the induced current of electricity cannot pass fluids, but probably because of its brief duration and feeble intensity; for on introducing two large copper plates into the circuit on the induced side (20.), the plates being immersed in brine, but prevented from touching each other by an interposed cloth, the effect at the indicating galvanometer, or helix, occurred as before. The induced electricity could also pass through a voltaic trough (20.). When, however, the quantity of interposed fluid was reduced to a drop, the galvanometer gave no indication. 24. Attempts to obtain similar effects by the use of wires conveying ordinary electricity were doubtful in the results. A compound helix similar to that already described, containing eight elementary helices (6.), was used. Four of the helices had their similar ends bound together by wire, and the two general terminations thus produced connected with the small magnetising helix containing an unmagnetised needle (13.). The other four helices were similarly arranged, but their ends connected with a Leyden jar. On passing the discharge, the needle was found to be a magnet; but it appeared probable that a part of the electricity of the jar had passed off to the small helix, and so magnetised the needle. There was indeed no reason to expect that the electricity of a jar possessing as it does great tension, would not diffuse itself through all the metallic matter interposed between the coatings. 25. Still it does not follow that the discharge of ordinary electricity through a wire does not produce analogous phenomena to those arising from voltaic electricity; but as it appears impossible to separate the effects produced at the moment when the discharge begins to pass, from the equal and contrary effects produced when it ceases to pass (16.), inasmuch as with ordinary electricity these periods are simultaneous, so there can be scarcely any hope that in this form of the experiment they can be perceived. 26. Hence it is evident that currents of voltaic electricity present phenomena of induction somewhat analogous to those produced by electricity of tension, although, as will be seen hereafter, many differences exist between them. The result is the production of other currents, (but which are only momentary,) parallel, or tending to parallelism, with the inducing current. By reference to the poles of the needle formed in the indicating helix (13. 14.) and to the deflections of the galvanometer-needle (11.), it was found in all cases that the induced current, produced by the first action of the inducing current, was in the contrary direction to the latter, but that the current produced by the cessation of the inducing current was in the same direction (19.). For the purpose of avoiding periphrasis, I propose to call this action of the current from the voltaic battery, volta-electric induction. The properties of the second wire, after induction has developed the first current, and whilst the electricity from the battery continues to flow through its inducing neighbour (10. 18.), constitute a peculiar electric condition, the consideration of which will be resumed hereafter (60.). All these results have been obtained with a voltaic apparatus consisting of a single pair of plates.

Section S19.
Estimated lower bound to the cost of materials using fluorescent Molbits: $2.4 \cdot 10^{-5}$ $ per GB.

Estimation of the lower bound of cost per GB
We estimate the lower bound of material cost (substrate, molecular inks, enclosure) for a cartridge holding a reel of a thin polymer film, which bears a maximum dense dot matrix of fluorescent Molbits (500 nm dot-to-dot distance), for digital data storage.
Our cost model is based on parameters given by the current generation of LTO magnetic tape (thickness and width of the polymer film; volumetric occupancy of the tape in the cartridge), estimated cost of components (cost of polyethylene terephthalate, PET; cost of the cartridge; cost of the dye), and a set of assumptions:

- Writing on both sides of the material.
- Leaving safety margins of 1 mm on each side of the film (84% of the total area).
- The amount of ink required is based on a spherical drop with the cross-sectional area of the projected minimum dot size (500 nm).

Derivation of the cost equation

symbol	description	quantity	value
$\$_{material}$	cost of all material	$/GB	calculated
$\$_{substrate}$	cost of the substrate	$/GB	calculated
$\$_{inks}$	cost of ink	$/GB	calculated
$\$_{dye}$	cost of dye	$/kg	100
$\$_{cartridge}$	cost of the cartridge	$/GB	calculated
$\$_{parts}$	cost of the parts to the cartridge	$/cartridge	2.0
d_{medium}	information density on the film ("volumetric information density")	GB/m^3	calculated
Q_{area}	occupancy on the medium	dimensionless	0.84
N_{col}	number of dyes/colors	dimensionless	8
N_{col}	number of sides written	dimensionless	2
r	center-to-center distance of spots	m	5.0×10^{-7}
d_{film}	thickness of the film	m	5.6×10^{-6}
c_{dye}	concentration of the dyes in the inks	g/L	0.1
$V_{cartridge}$	volume of an LTO-9 cartridge	m^3	102 mm × 105.4 mm × 21.5 mm = 0.00023 m^3
Q_{area}	occupancy of the film in an LTO-9 cartridge	dimensionless	0.33

$$\$_{material} = \$_{substrate} + \$_{inks} + \$_{cartridge}$$

$$\$_{substrate} = \$_{polymer} \cdot \rho_{polymer} \cdot \frac{1}{\delta_{medium}}$$

$$\delta_{medium} = \frac{\theta_{area} N_{col} N_{side}}{8 r^2 d_{polymer}}$$

$$\$_{inks} = \$_{dye} \cdot c_{dye} \cdot r^3 \frac{\pi}{6}$$

$$\$_{cartridge} = \$_{parts} \cdot \frac{1}{\delta_{medium} \cdot V_{cartridge} \cdot \theta_{cartridge}}$$

$$\$_{material} = \$_{polymer} \cdot \rho_{polymer} \cdot \frac{8r^2 d_{polymer}}{\theta_{area} N_{col} N_{side}} + \$_{dye} \cdot c_{dye} \cdot r^3 \frac{\pi}{6} + \$_{parts} \cdot \frac{8r^2 d_{polymer}}{\theta_{area} N_{col} N_{side}} \cdot \frac{1}{V_{cartridge} \cdot \theta_{cartridge}}$$

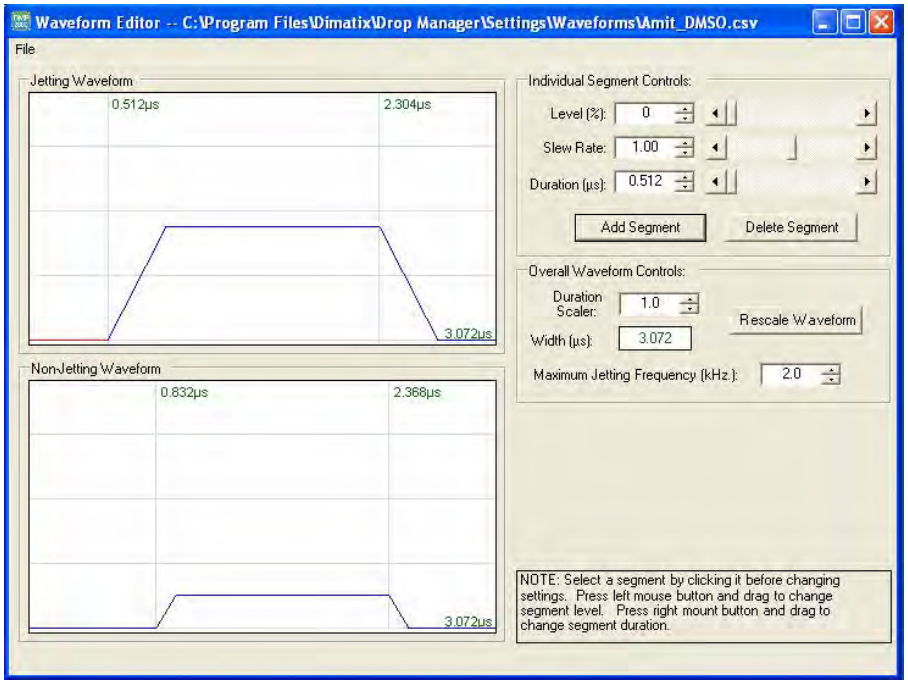

Figure S20. Typical jetting and non-jetting waveform used to ink-jet print DMSO solutions of the fluorescent dye molecules. The voltage for jetting of the piezoelectric nozzles was 20V.

Section S21. Estimation of the lifetime of fluorescent dyes

In this report, we use commercially available fluorescent dyes synthesized for tracking biological events. Many of the dyes are substituted fluorescein dyes. Fluorescein-based dyes are used as trackers in geothermal reservoirs and its decay kinetics have been studied in detail.

The first order decay parameters for fluorescein in distilled water are available in literature (Geothermics 1991, Vol. 20, No. 1/2, 53-66)[1] where:

Arrhenius equation:

$$k = Ae^{-E_a/RT}$$

$$\ln k = \ln A - \frac{E_a}{RT}$$

For degradation of fluorescein in distilled water:

E_a = activation energy = 163880 J/mol (= 39.2 Kcal/mol) and

ln(A) = 22.87 where *A* = Arrhenius frequency factor.

Hence, the rate constant *k* at room temperature (298K) is: $1.6 \times 10^{-19}\ s^{-1}$

Using these calculations at room temperature (298K), the time required for 10% reduction in concentration of fluorescein in distilled water is:

First order reaction kinetics: $C = C_0 e^{-kt}$,where C/C_o = 0.9 for 10% reduction in concentration.

Where *C* = concentration of fluorescein remaining at time *t*, and C_o = initial concentration of fluorescein. Hence, t for 10% reduction in concentration is:

$$t = -\frac{1}{k} \ln \frac{C}{C_0}$$

= 6.56 x 10^{17} seconds = (approximately) 2 x 10^{10} years.

While this number is very large, substituents on the fluorescein molecule will change this lifetime considerably. This large timescale demonstrates the fact that fluorescent molecules can be very stable at ambient conditions.

References

[1] M. C. Adams, J. Davis Geothermics **1991**, *20(1/2)*, 53-66.