Supporting Information for:

Fast, energy-efficient synthesis of luminescent carbon quantum dots

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S1: OTHER COMBINATIONS



Figure S1-1: Luminescence of C-dots synthesized by a 4 saccharide/base combinations. Dialysed for 24 hours, 1 week after mixing solutions.



Figure S1-2.Photoluminescence spectra of other saccharide and base mixtures.Spectra collecteduisngtheHitachiF-700

S2: DECONVOLUTION OF RAMAN SPECTRA:

- C-dots, peak deconvolution from 1200 1500 cm⁻¹ from Figure 4(a)
- This fit was done using a Gaussian fit in Origin Pro 9.0



Figure S2. Deconvolution of peak in Raman spectrum from Figure 4 (a).

The labelled D peak from Figure 4(a) upon peak deconvolution of the area (and the near fit of the cumulative peak function (blue dot-dash), yields 2 peaks exist around 1357 cm⁻¹ (red dot-dash series – attributed as the standard D peak) and 1418 cm⁻¹ (green dot-dash series).



Figure S3. Labeling of peaks in FTIR spectrum from Figure 4(b)

Peak [cm ⁻¹]	Probable Assignment	Ref
3430	O-H (stretch)	R1-R5
2930	C-H (stretch)	R1,R4-R6,R8
2855	C-H (stretch)	R4,R6
1650	C = C (stretch)	R2
1380	C-H (bending, -CH ₃ functional group)	R1,R7
1110	C-0	R2
1050	C-0	R1
~881	C-H bend	RF8

<u>S4: LONG TERM STABILITY OF C-DOTS</u>



Fig. S4-1: Visible luminescence upon excitation by 405 nm laser of samples taken 1.5 years after synthesis



Fig. S4-2: Visible luminescence upon excitation by 405 nm laser of samples taken 10 months after synthesis.

S5: PLAUSIBLE FORMATION MECHANISM

What is the effect of concentration of solutions on the reaction kinetics?

From the experiments described in the main text, we know that if concentrations of saccharide and base are not high enough, then C-dots will not form without extra energy input into the system, be it via external heating or applied voltage. The energy to ensure C-dot formation takes place, must come from somewhere – thus the only viable candidate in this situation would appear to be the mixture itself, which suggests that we should consider the Nernst Equation.

Nernst Equation:

 $E_{cell} = E_{cell}^{o} - \frac{RT}{nF} \ln Q$

where E_{cell}^{o} is the standard cell potential at 1M concentrations, *R* is the gas constant, *T* is the temperature, *F* is Faraday's constant, *n* is the number of electrons being transferred in the redox reaction and *Q* is the reaction quotient ^{R9}

$$Q = \frac{[reduction]^{y}}{[oxidation]^{x}}$$

If we consider that the reaction is just occurring at room temperature, the following form can be used:

$$E_{cell} = E_{cell}^{o} - \frac{0.0592}{n} \log Q$$

Recall, $\log_{10}(a/b) = \log_{10}a - \log_{10}b$, therefore (< means decrease, > means increase):

[Red] [Ox]	Same	Increase	Decrease
Increase	$\log Q <, E_{cell} >$	-	-
Decrease	$\log Q >, E_{cell} <$	-	-
Same	No change	$\log Q >, E_{cell} <$	$\log Q <, E_{cell} >$

Now consider what species we have in the reaction vessels, i.e. what species are available in the combined volumetric flask to make up the C-dots?

Moreover, which act as the reductant and which as the oxidant?

But firstly ... which combination of sugar and base work?

The formation of C-dots in this case is likely a redox process, which rules out (unaltered) sucrose as a suitable starting material as it is not a reducing sugar. Maltose, the other disaccharide considered is a reducing sugar as it has an aldehyde group, as does glucose. Fructose is also considered a reducing sugar. Sodium hydroxide (NaOH) is a strong base, whereas sodium bicarbonate (NaHCO₃) is a weak base

	NaOH	NaHCO ₃	Reducing
Fructose	Y	Y	Y
Glucose	Y	Ν	Y
Maltose	Y	Y	Y
Sucrose	N	N	N

Figure S1: Results table for various sugar/base combinations

• What is happening in the reaction vessels?

In beaker 1 (Scheme 1 of the main text):

A disaccharide, such as maltose, upon addition of H_2O may undergo hydrolysis – breaking up into 2 glucose molecules (note, as it contains an aldehyde group it may also participate in the redox process without breaking up into monosaccharide units)

$$C_{12}H_{22}O_{11} \text{ [maltose]} + H_2O \rightarrow C_6H_{12}O_6 \text{ [glucose]} + C_6H_{12}O_6 \text{ [glucose]}$$
(1)

The glucose/fructose may then undergo the next reaction, however, this typically requires a catalyst, so is less likely in this synthesis scenario, but not impossible:

$$C_6H_{12}O_6 + H_2O \rightarrow C_6H_{12}O_7 + H_2$$
(2)

In beaker 2 (Scheme 1 of the main text):

The sodium hydroxide or sodium bicarbonate will dissociate upon addition of H₂O and will release heat.

$$NaOH + H_2O \rightarrow Na^+ + OH^- + H_2O + heat$$

$$NaHCO_3 + H_2O \rightarrow Na^+ + HCO_3^- + H_2O + heat$$
(3)
(4)

The HCO_3^- may undergo a further reaction with H_2O :

$$HCO_3^- + H_2O \rightarrow H_2CO_3 + OH^-$$
(5)

In combined volumetric flask:

Likely, the following species will be present and available to undergo reactions which will contribute to C-dot formation: Ions: Na⁺, OH⁻, HCO₃⁻

 $\textbf{Molecules: } C_6H_{12}O_6, H_2O, NaHCO_3, NaOH, H_2CO_3, C_6H_{12}O_7, H_2$

Therefore, the amount of heat depends on the concentrations of the bases that are added to the hydrocarbon precursors. This is consistent with the results of our experimental observations mentioned in the Experimental Section of the main text. Further decomposition of hydrocarbon precursors will lead to the formation of building units (species) needed for the nucleation and growth of the C-dots.

Regarding stabilisers and surfactants:

The interesting property of C-dots is their outstanding solubility and dispersion in aqueous solutions. This was clearly seen in our experiments, without any additional surfactants or stabilisers. This feature is very different from carbon nanotubes with the very different (namely water repellent) surface chemistry which leads to the conclusion that the (intrinsic) surface chemistry of the C-dots attained without any surfactants makes them reasonably hydrophilic and soluble. Since the saccharide molecules are reformed into quantum dots (near all or all reagents eventually react - **Note:** It is possible to obtain C-dots without waiting a week for dialysis (see Figure 1 – main text, for example, re: the visible luminescence). However in this case the week long wait was taken as a precaution to ensure that everything that could react did, i.e. we waited until there was no further colour change observable) it is unlikely that un-reacted saccharide molecules act as surfactants. Moreover, the C-dots are very stable in the solution, even after 15 months, see ESI, Section 4. The mechanism of this stability is likely related to the structural integrity and crystallinity of the C-dots and further studies are required to confirm this assumption.

S6: NOTES ON TOXICITY EVALUATION AND POTENTIAL BIO-APPLICATIONS

An assay-based assessment is required when a biological application is intended. In our case, since the emission is in the green range, the intended application may not be primarily aimed for bio-imaging. This is why the toxicity evaluation is of less relevance in this case. As we will continue our work, we aim to eventually achieve spectral emission in the NIR/IR range of particular importance for bio-applications (see our response to the previous comment); that study indeed envisages the cytotoxicity evaluation.

More information on C-dot toxicity can be found elsewhere [R10].

S7: REFERENCES

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