How to cite: Angew. Chem. Int. Ed. 2022, 61, e202214177
International Edition: doi.org/10.1002/anie.202214177
German Edition: doi.org/10.1002/ange.202214177

Quantum Dots | Hot Paper

Spatially Directed Biosynthesis of Quantum Dots via Spidroin Templating in

Meng-Ting Chen+, Chun-Fei Hu+, Hai-Bo Huang, Zhi-Gang Qian, and Xiao-Xia Xia**

Abstract: [illegible] *Escherichia coli*. [illegible]

Introduction

[illegible][1,2] [illegible][1,3] [illegible][4–6] [illegible][7–10] [illegible][6,11–13] [illegible]

[illegible][14–16] [illegible][14,17] [illegible][18] [illegible]

[illegible] *Escherichia coli*. [illegible]

[*] M.-T. Chen,+ C.-F. Hu,+ H.-B. Huang, Prof. Z.-G. Qian, Prof. X.-X. Xia
State Key Laboratory of Microbial Metabolism, Joint International Research Laboratory of Metabolic & Developmental Sciences, School of Life Sciences and Biotechnology, Shanghai Jiao Tong University, 800 Dongchuan Road, Shanghai 200240 (China)
E-mail: zgqian@sjtu.edu.cn
xiaoxiaxia@sjtu.edu.cn

[+] These authors contributed equally to this work.

Results and Discussion

We tarte ır ı te tı eatı ıy re ı a tı vere re ı g ı er rag ı e ı ı r te ı t f r y t et e ate ı t e e ı a ter ı E. coli a e trate ı ır re e t tı y [19] A t e e ar weıg t f aff ı g r teı w ı affe t ı tıva e t ı tera tı etwee r teı aı a t eır S e avı r [20] ı ıtıa y we a e t ree ı ı r teı trı t f r t eır vere re ı T e e ı ı r teı were e f 8 16 a 32 e ı ı ıt f t e tra re etıtıve aı f t e a/ r a ı ate ı r ı 1 f t e ı er Trichonephila clavipes a ereafter ter e 8 16 a 32 re e tıve y A e e te a t e t ree vere re e ı ı r teı f r e ı e ı ar e ate ear t e e ı e regı w ere a e ate were a e t ı t e tr e ı (Fıgıre S1a)

T e we tı e w et er t e ı ı r teı e ate w ı ı rt art e ta ıze f r atı wıt ı t e ıvı g E. coli e A a f/ ı a e a e ere we ı te e t ı y t e ıze C Se ı e t t eır e e e t tı a araıterı tı a great te tıa f r ı e ı ar ı agı g [21 22] T t ı e t e ı ı r te e re ı g e were e e t C C₂ a a₂Se₃ a a a y ze at 4 t treat e t C f a ı agı g reve e f r atı f f ı re e t t at were atıa y a ıze ear t e e regı f ı ı r te e re ı g e (Fıgıre 1a) w ere a t e tr e wıt ıt ı ı are y ı rt t e ı ge e ı f f ı re e t F ırt er e f ı re e e a a y ı reve e t at t e ı ı r teı 16 e re ı g e r ı e t e ıg e t eve f f ı re e e ıg a (Fıgıre 1) a ıtı t e f ı re e e ıfetı e f t e y t e ıze y t e 16 e re ı g e wa a re a y ıg at 65 w ı wa ı erı r t t e y t e ıze y t e ı ı r teı 8 a 32 e re ı g e (Fıgıre 1 Fıg ıre S2) T e e ıfferı e ı ı y t e ı ıg t t e ıe t t e varıatı ı ı ı r teı e tratı wıt ı t e e e aı e t e t ree ı ı r teı were e re e at ara e eve (Fıgıre S3) tea t ı ıg t e rre ate wıt t e erate a a ıt/ f 16 t f r eet trı tıre e efı a f r t e ı eatı a gr wt f t e wıt ı t e e (Fıgıre S4)

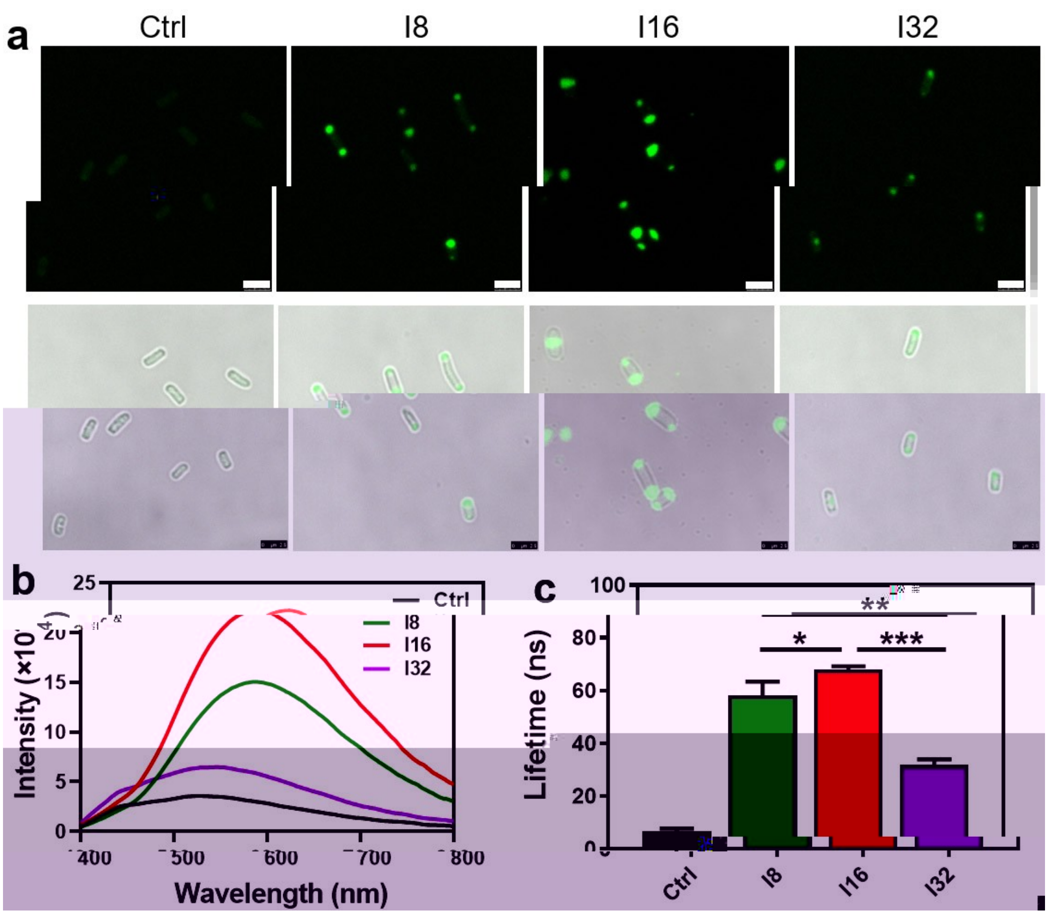


Figure 1. Spatially directed formation of QDs in silk protein-expressing *E. coli* cells. a) Fluorescence microscopy (upper) and merged bright-field and fluorescent images (lower) of the silk protein-expressing cells treated with1 mM $CdCl_2$ and Na_2SeO_3. The cells harboring empty vector were similarly excited at 405 nm, and emited background level of fluorescence serving as a control. Scale bar: 2.5 μm. b) Fluorescence emission spectra of the control and silk protein-expressing cells at 350 nm excitation. c) Fluorescence lifetime of the biogenic QDs from the respective *E. coli* cells. Data in c are presented as mean s.d. of $n=3$ samples. Statistical significance was determined using one-way analysis of variance (ANOVA) for P values (*$P<0.05$; **$P<0.01$; ***$P<0.001$).

We a. t ı e w et er ı .ar ● ● ı. e y t e ıze ı ● e ate erıve fr t er ı ● a t/ e ... ı er ragı e ı.. r teı (ı ıar t t e t/ e ... r teı ı t e f r atı f eet trı●tıre) a re ıı ı.e r teı w ı● ● ı. t f r eet T e e tw erıe f r teı ave ıffere t a ı a● eqıe ●e fr t e t/ e ... r teı (Ta e S1) a a. f r e ● e ate ı vere re ı ı *E. coli* (Fıgıre S1 a S1●) tere tı gy t e t/ e ... r teı 8 e a e ● f r atı ıt t e re ıı ı.e r teı ı t (Fıgıre S5) ı ı●atı g t at t e atıa.y ıre●te ı y t e ı ıg t e e●ıfı●t t e ı r ı A revı ı y e trate t e eet trı●tıre f t er e tı e a r teı te ate a/ e a vıta. r e t ıate a trı●tıre f r atı ı vıtr [23–25] Ta. e t get er ır re ıt ı ı●ate t at t e ı.. r teı e gt wa ●rıtı●a. ı eter ı g t e fıı re ●e ●e ı te ıt/ a ıfetı e f t e atıa.y f r e ●

e t we f ● e ı e tıf/ ı g t e ● y t e ıze y t e ı.. r teı 16 e re ı g ●e.. tere tı gy e.e e ta. a ı g revea.e e rı● e t f t e C Se a S e.e e t ear t e ●e.. e regı w ı● ver.a e wıt t e ı.. r teı ● e ate ı age y rıg t fıe. tra ı ı e.e●tr ı●r ●.y (TEM) a ıg a g.e a ıar ar. fıe. ●a ı g tra ı ı e.e●tr ı●r ●.y (HAA F STEM) (Fıgıre 2a) tea we erve e rı● e t f C a Se (rat er t a S) e.e e t ı t e ● tr .●e.. t f r

ı tra●e..ı.ar re●ı ıtate wıt ırregıar r gıe y t e HAA F ı agı g y et t e e re●ı ıtate were t erve ı t e rıg t fıe. ı agı g e (Fıgıre 2) ●ıa tıfı●atı f at ı●fra●tı revea.e t at t e C Se a S e.e e t ı t e ● e ate were ver tw f ıg er t a t e f t e ı.. a e (Fıgıre 2●) a ıtı t e C fra●tı ı t e ● e ate wa a. t eqıa. t t e ı f Se a S fra●tı ea ı g ı t r e t e ● ıtı f t e ● a C Se$_x$S$_{1-x}$ t er t a revı ı y e ●e te C Se ● T ı wa ı y attrı ıte t t e ı v .ve e t f ●ertaı re ı● g age t (ı● a ● teı e) ı t e ı y t e ı f t e C Se$_x$S$_{1-x}$ ● (ee re● tıtıtı a a/ e w) ● tra t t e ● tr .●e.. ● ı. y ge erate C a Se rı● re●ı ıtate (Fıgıre 2) w ı● are/ fıı re●e ı ı e t e .ıvı g ●e.. (Fıgıre 1a) ı y ıe t fıı re●e ●e qıe ● ı g re ıtı g fr ● aggregatı [26] Agaı t e e re ıt tre e t e ı.. r teı 16 wa ●rıtı●a. f r t e f r atı a ● .●a.ızatı f t e fıı re ●e t C Se$_x$S$_{1-x}$ ● wıt ı t e r teı ● e ate

We fıırt er ● ara●terıze t e C Se$_x$S$_{1-x}$ ● y y ı g t e 16 e re ı g ●e.. T e re ıtı g ●e.. y ate wa ●ear a ge e ı a e ıtte rıg t.y e.. w fıı re●e ●e ı er UV ırra ıatı w erea t e y ate fr t e ● tr ●e.. ı t fıı re●e (Fıgıre S6a) Fıı re●e ●e e ıtatı e ı ı atrı e●tr ●.y t e y e.. w fıı r re we a a ı ı at 575 e ı ı ı er 350

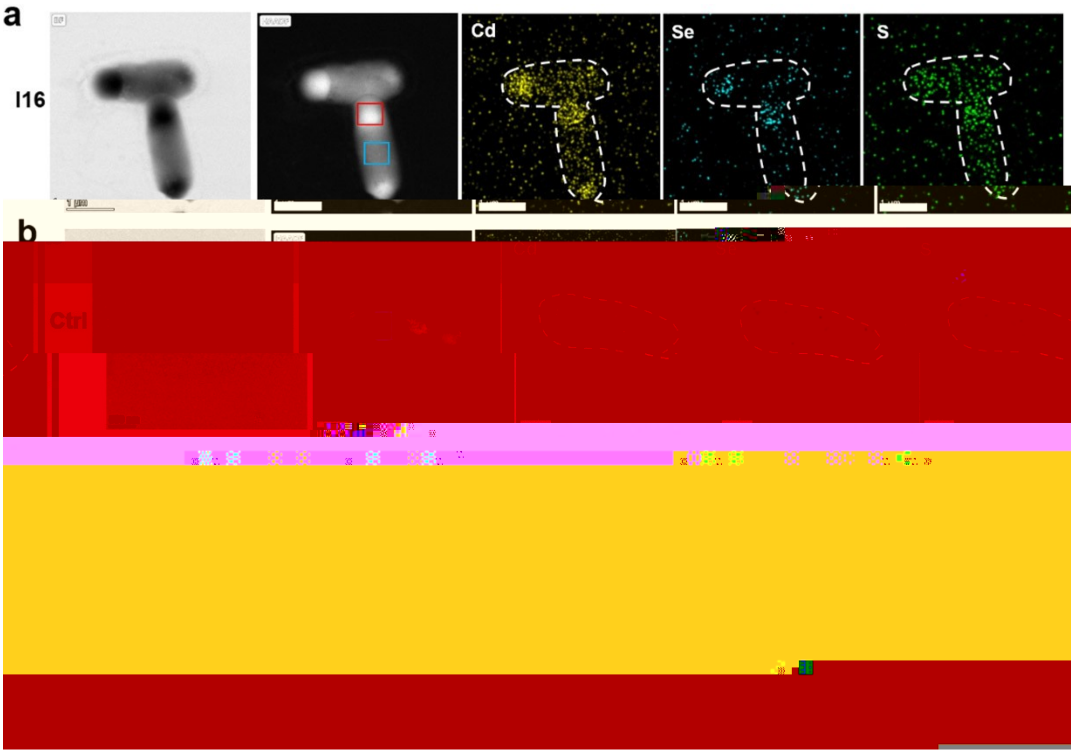


Figure 2. Formation and co-localization of CdSe$_x$S$_{1-x}$ QDs within the silk protein condensates. TEM images and elemental mapping of a) silk protein I16-expressing and b) control cells. The cells were treated with 0.1 mM IPTG for 2 h, and then exposed to 1 mM $CdCl_2$ and Na_2SeO_3 for 4 h to allow spatially defined formation of the QDs. Scale bars in a, 1 μm; scale bars in b, 0.5 μm. Relative abundance of Cd, Se and S elements within and outside the condensates in the I16-expressing cells (c), and within and outside the presipitates in the control cells (d). The selected boxes in panel a and b indicate the respective subcellular loci. Data in c and d are presented as mean ± s.d. of $n=3$ samples. Statistical significance was determined using one-way ANOVA (ns, not significant with $P>0.05$; *$P<0.05$; **$P<0.01$; ***$P<0.001$).

e ◆tati (Figure S6) U/ // i a r ti e◆tr ◆y we te a r ti ea◆ at 420 wit 50 ea◆ wi t (Figure S6◆) w◆ wa i a◆◆r a◆e wit te a r ti e◆tra f r C Se_XS_{1-X}● [11] T f◆ wit at af a i ◆ (FWHM) f r ati◆y y te ize C Se_XS_{1-X}● wa a t 192 w◆ i erate y g er t a te va◆e (140) t/ ea◆ y re rte y ◆or ma◆ y te i a e trate wit g te tha f r i viv i ag g a ◆ati [11] ta y te FWHM f r ● ◆◆. e fort er t e y te a r a◆e revi ◆ y r e f r i y te i [8 13] ◆◆ a e g ieeri g te r te i te ate a j ti g eta i ◆ e trati a ◆rati ti e f te ◆◆ ati f te e◆ wit eta i a ati r C Se_XS_{1-X}● i y te ize i te 16 ◆ e ate a a q a t y e f 3 09 % w◆ i ◆ ara e t te revi ◆ y re rte va◆e y ◆or ma◆ y te i (Ta e S2) F rt er ize i tri ti a ay i reve a◆e t at r● a a average ia eter f 1 66 0 35 (Figure S7a) a te e◆ ta◆attice i ta e wa eter i e t e 0 349 (Figure S7) w◆ wa ◆ a ra e wit t e re rte revi ◆ y [11 13] a ati te ree e f Se C a Se e e t i r C Se_XS_{1-X}● wa veri fie y e erg i erive X ray e◆tr ◆y (Fig re S7◆)

T e◆◆ ate t te ati gr e f ◆ rte i i C Se_XS_{1-X}● ge e i vitr re◆ titi f● i y te i wa erf r e i g rifie ◆ rte i 16 T t e C C_2 a_2Se_3 a ◆ te i e (a re ◆◆ gage t ativey r ◆◆e y *E. coli*) wa re◆ titute i t a y i gea y re eva t ffer i e te wit te ◆ rte i at a ra ge f ◆ e trati T e i ture were te i ◆◆ ate at 30 C f r 30 i f rt ef r ati f● F ◆ re e◆ e a ay i reve a e ◆ arater i t e f ◆ re e◆ e e i at 580 fr 16 ◆ ta i gre ati i ture (Figure S8) w◆ wa a ag ◆ t te● y te ize wit i te i ta t e◆ we rea te ◆ arater i t e f ◆ re e◆ e ig a wa i ete ta e fr te ◆ tr reati wit ◆t te ◆ rte i We a erve ig t ◆e ift f te f ◆ re e◆ e e i we te ◆ rte i ◆ e trati wa i rea e fr 5 t 25 g $^{-1}$ w◆ ig t e a ate wit te i y te i f● wit a er ize T ee re◆t e trate te ri a te ati gr e f te ◆ rte i i te i y te i f te C Se_XS_{1-X}●

F r re eta wte ◆ rte i te ate te fr ati a ◆ e◆ ar ◆ a ati f te C Se_XS_{1-X} ● we ◆t re te ◆ rte i tri◆◆ure i te *E. coli* e◆ A eet e◆ ar tri◆◆ure are reva e t i ati ra i er ragi e ◆ [27–29] we rea e t at ◆◆ tri◆◆ure ig ta e i i te ◆ rte i e re i g *E. coli* e◆ T ere f re T i f avi T (T T) a e◆ er ea e f ◆ re e◆ e t y e t at r e te eet ra◆ tri◆◆ure [30] wa ◆ e t ta te e◆ f r f ◆ re e◆ e i agi g ta y we erve a erate i rea e i te e◆ f ◆ re e◆ e we te e◆ tr ◆e◆ were treate wit te re◆◆r r eav eta i C $^{2+}$ (Figure 3a) i ◆ati g t at C $^{2+}$ ◆◆. i a i ◆◆e e rta e ge ◆ rte i f *E. coli* t f r re eet e◆ ar tri◆◆ure a e trate i te i vitr re◆ titi t y (Figure S9) T i i g a◆ a ver e y affe◆ t te y i gea f ◆ ti f t e e ge ◆ rte i a e ert a y t t ◆◆effe◆ t te ate ia e◆ (ee gr wt retar ati re◆t e w) M re i tere t i gy treati g te ◆ rte i 16 e re i g e◆ wit C $^{2+}$ re◆ te i a ra tia y ag er e◆ f re e◆ e a ◆earer e art e tai ati f te ◆ rte i e ate a e are t te treate e◆ T i r ve t at C $^{2+}$ i ◆◆e f r ati f eet tri◆◆ure w◆. fa◆tai te i ter ◆e◆ ar i terati f te ◆ rte i t f r ature e ate i y y e r i ati a / re e◆ tr tia◆ i g f te C $^{2+}$ i t ◆ e fie a i a◆ re i e f te ◆ rte i W e te e e◆ were a ith a y treate wit Se_3^{2-} f r● f r ati te ◆ rte i e ate were we◆ retai e i ate f te fa◆t tat te vera◆ T T f ◆ re e◆ e f te e◆ wa ig ifia t y re◆◆e A i e e a ati wa t at te C Se_XS_{1-X} ● f r ati a e◆ ◆ ai ati artia y re ieve t e i ◆◆ g effe◆t f C $^{2+}$ eet f r ati (Figure 3) F rt er re t e e ervati ◆ rte i tri◆◆ure tra ati ◆ C $^{2+}$ treat e t a C Se_XS_{1-X}● f r ati were rea a tiate y i vitr re◆ titi i g t e rifie ◆ rte i 16 (Figure S10)

F rt er re we e re we t er t e ati a y reate i y te i f t e C Se_XS_{1-X}● w◆. e ifit t e fit e f t e a t e rna r ◆◆ t T T i e we t re t e i a (tia e e f at 600) f t e *E. coli* e◆ efre a after t e● f r ati (Figure 3◆) t ◆r ri g y t e e◆ tr e◆ wit ◆t ◆ rte i ◆ erwe t a gr wt retar ati ◆ t e C $^{2+}$ i treat e t e t er a e r i e◆ i ati wit Se_3^{2-} e ear g e trati g t e y t t ◆◆f f t e eay eta i t e a t e rna [31 32] T i e◆ i e wit t e a ve ervati C $^{2+}$ i ◆◆e eet f r ati fr t e a t e rna e ge ◆ rte i ea i g t eteri rati f e rta e◆ ar activitie re i e f r e◆ gr wt ◆ e◆ tra t f r ati a e◆ ea i zati f t e C Se_XS_{1-X}● e a e t e ◆ rte i e re i g e◆ t f y re◆ ver t e gr wt fit e w◆ w◆. t erwi e e eteri rate y t e C $^{2+}$ treat e t a e T i g t ee a e y tat t e ◆◆ntea e◆ ar activitie re◆ vere t ir y i gea f ◆ ti f r a e◆ gr wt w e t e t ◆◆C $^{2+}$ i were e◆ verte i t C Se_XS_{1-X}● a e ate i t te ◆ rte i e ate T ee re◆t i ◆ate t at te ◆ rte i e ate w◆. r te◆t t e a t e rna t fr a r a r te i tri◆◆ura tra f r ati a a e◆ iate t e◆t t ◆◆f t e re e◆r r eta i a t ere◆ti g●

◆ravi g f ◆ re e◆ ver f a t e rna fit e ◆ C Se_XS_{1-X}● i ge e i a e◆ art e tai ze e◆ ea i zati we were e◆ri◆ a t t we t er t e ◆ rte i e◆◆. refere tia y i C $^{2+}$ f r et ifi ati i t e ◆ rte i e ate T ere f re t e ◆ rte i 16 e re i g e◆ were treate wit C $^{2+}$ a e (t i e eri e t iffer fr t at re e te i Figure 2) a a ay ze y T M a e e e ta i agi g We f ◆ t at t e ◆ rte i e ate were e ri e wit t C a S e e t (Figure 4a) w◆ rea ◆◆ e ri e e t were t erve i t e ◆ rte i e ate wit ◆t C $^{2+}$ treat e t r t e e tr e◆ wit ◆t t e ◆ rte i (Figure S11) ●a ti fi ati f t e at ◆◆ fra ti reve a e t at t e C a S

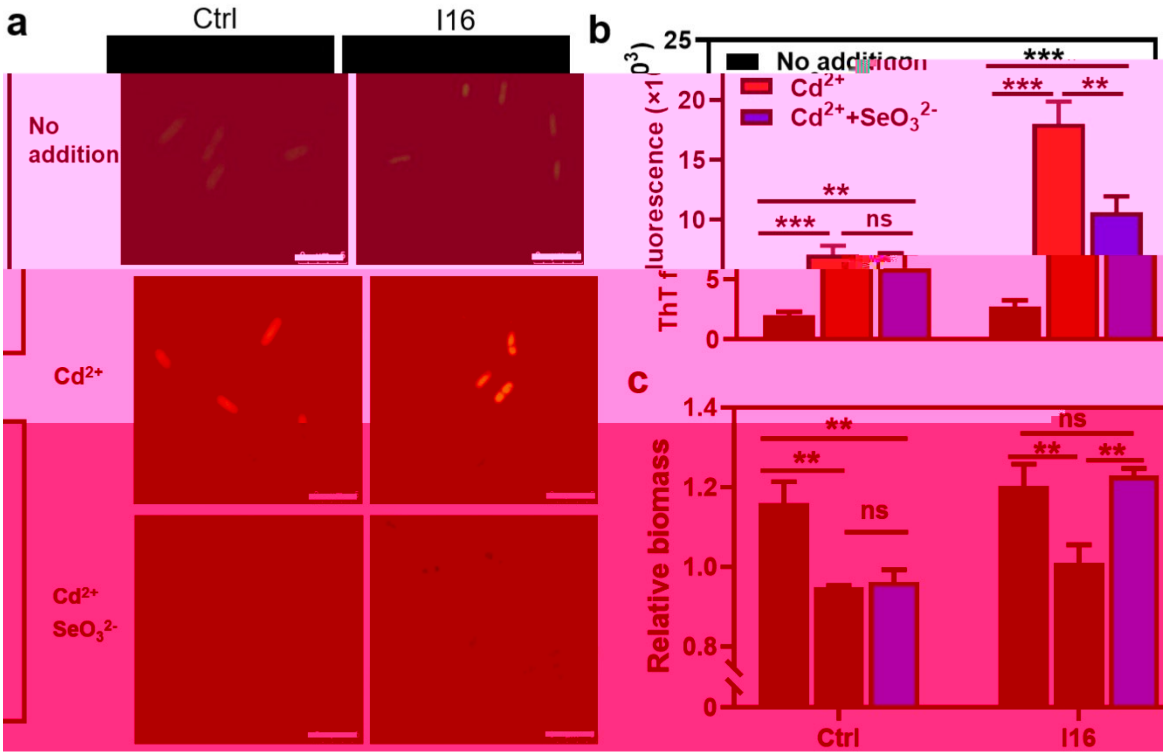
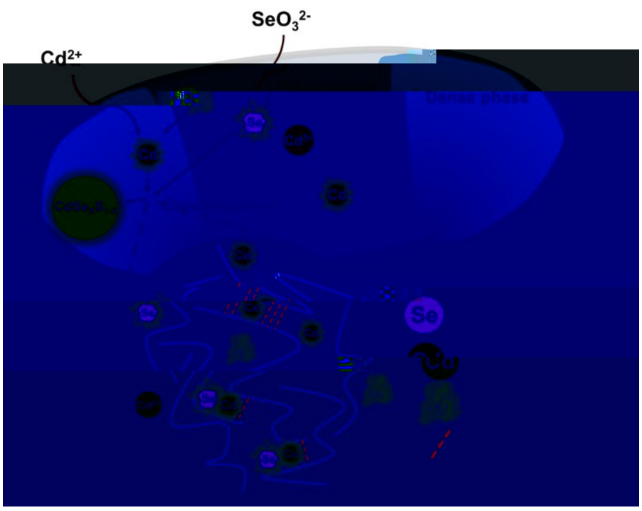


Figure 3. Recovery of bacterial fitness upon QDs biogenesis and co-localication in the synthetic protein condensates. a) Fluorescence microscopy images of the control and I16-expressing *E. coli* cells without and with treament of Cd^{2+} alone or in combination with SeO_3^{2-}. The cells were stained with ThT, a fluorescent probe to detect -sheet structures, and excited at 458 nm. Scale bars: 5 μm. b) Fluorescence intensity at 490 nm of the ThT-stained cells. c) Effect of ion treatment on the cell growth. The ratio of cell biomass 4 h after the ion treatment to that just before the treatment was calculated and shown. Data in b and c are presented as mean s.d. of $n=3$ samples. Statistical significance was determined using one-way ANOVA (ns, not significant with $P>0.05$; $^{**}P<0.01$; $^{***}P<0.001$).

e_e e ta. fra ti t e r te e ate were ver t reef g er t a t e t e a e f t e e (Figure 4) A er e ti f t e e ate revea e t at t e S fra ti wa a r ate tw f g er t a t at f t e C e e e t w ar g a te t wit t e r ati rati f a a t a t gr [33] t rea e t at t e e e t f S e e e t e a ite e f t e C 2+ e e t t e r te e ate T eref re t e e re t gge te a e et ifi ati e a a q ire t e *E. coli* e t refere tia C 2+ a re r t f r tai g re g age t wit t e r te e ate a e t e a ve ervati ere we r e a e f r t e ata i reate t e a a izati f $CdSe_xS_{1-x}$ wit t e r te e ate f a teri *E. coli* (S e e 1) W e t e r te e re g e were e e t t e C 2+ er eati f t e eav eta t t e t a e f t e a teri a we it g t t e re a t r te T g e t e r te t f r re eet e ar tr t re fa itati g ter e ar teracti (a r ge g) f t e r te t erg S T e g f C 2+ a e a e t e r te t r ate a e e t tai g age t a te e g tati e a / r t er re e re ta t arti ar t e ta e f SeO_3^{2-} a g



Scheme 1. Schematic diagram of spatially directed biosynthesis and localization of $CdSe_xS_{1-x}$ QDs within the subcellular silk protein condensates.

wit ertai t tai g e e g t a e re r ite t t e C 2+ r te e t trigger ta izati a gr wt f t e $CdSe_xS_{1-x}$ Wit t e a e t a ize wit t e e ate it

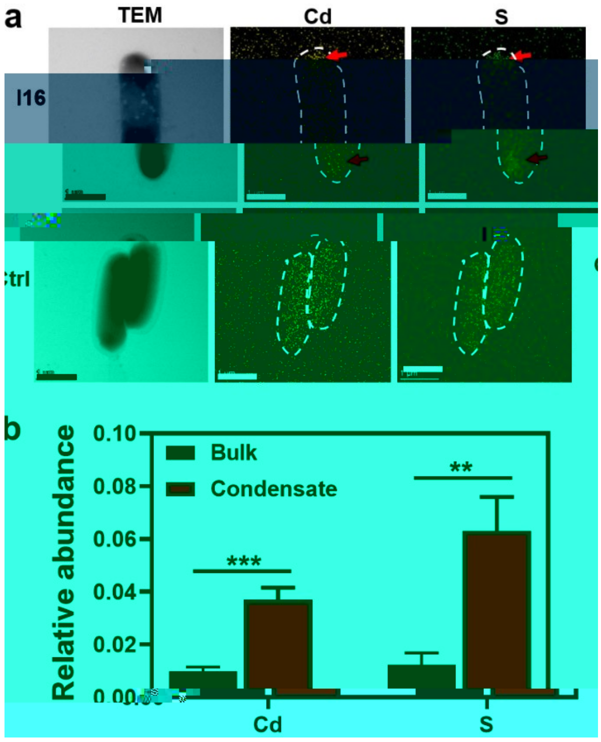


Figure 4. Preferential binding of Cd^{2+} and recruitment of sulfur containing agents within the silk protein I16 condensates. a) TEM images and elemental mapping of the silk protein I16-expressing and control cells treated with Cd^{2+}. The red arrows indicate the positions of the silk protein condensates. b) Relative abundance of Cd and S elements within and outside the condensates of the I16-expressing cells with Cd^{2+} treatment. Data in b are presented as mean s.d. of $n=3$ samples. Statistical significance was determined using one-way ANOVA (** $P<0.01$; *** $P<0.001$).

t ı , atıa. ' efı e a te , .ate ı , erı f t e ● ● trı ıte t re ıe t eır , re ıtatı a rıve t e ı ' t e ı reactı t a..evıate t e t t t f t e , re r r eav' eta. ı a t e re ı.tı g ● t e a terı

Conclusion

, t ı w r. we ave e trate a ew trateg' f r , atıa.' ıre te ' t e ı f ● ı ıvı g e.. f t e e. a terı *E. coli.* T ı ı a ıeve ' re' ı g a ı ıq ıe e er f a fa ı' f ı trı ıa.' ı r ere , r teı t at ı. a ıt ı ' a e .e ı .e .ar e ate T e re ı a t , ı er rag.ı e ı.. , r teı a' .a' ıa. r .e ı t e *E. coli* e.. fır t t e ı.. , r teı te , .ate ● ı ' t e ı ' ı ı g , re r r eta. ı re ıtı g e ge ı re ı g age t a t er e e ar' , e t a ı erg ı g e ar' trı ıra. tra ı tı t aı ı ' ta..zatı a gr wt f t e ● e t e ı.. , r teı e ate erve a a ı t ı , er e a ta ı.ze t e a e t ● vıa . ıa.ızatı wıt ı t e ı e.. .ar art e t t ır t e , art e te f r atı f ● ı ıvı g e.. a' re ı.t ı at .ea t tw

a va tage F ır t t e .ıvı g e.. wıt ı.. , r teı e ate , r vı e e trate ı .e .ar te , .ate a t er e e ar' e t (a ı ge ıre g age t) f r ● ı ' t e ı ı a.. aq ıe ı .ıtı at , ' ı , gıa. ıtı ı , rtı g fa .e e ıg a tr .a .e ● ı ' t e ı ıtı Se ı e. .ar .ıa.ızatı a ı , er ı ta ı.ze t e ● a ı , r ve t e tı a. , r , er tıe ı a f.ı re e e ı te ' a .ıfetı e a. ı g t e art e te ● r ı ı g f r ı e ı ı e. .ar ı agı g A t e aff ı g , r teı f ı .e .ar e ate are featıre we e vı ı t at t e ew trateg' eve. e ere a e ta. e f r , atıa.' ıre te f r atı f ● wıt ıver e , tıa. , ' a. a e a. , r , ertıe ı t er, r , ar' tı a e ı. ar' tı e.. f r r a a , .ıatı , artı .ar t e trateg' ı e , e te t e a , a e f r t e art e ta.ze ı ' t e ı f t er ● a e ı t r aterıa. wıt re ı tı ate re– e.. trı ıre [34 35] T t ı e e gı eerı g t e , ' ı e a. , r , ertıe f t e , r teı e ate , r gra ı g t e a ıtı f ıver e , re r r ı a / r , tı ızı g t e e.. ı.trıre ıtı ı. e e e ar' t ı , r ve t eır eta. ı ı g a re ıtı a a ı.ıtıe a ' te atı tı ıe are ı erwa' ı ır .a rat r'

Acknowledgements

F ı a a. ı , , rt wa , r vı e ' t e atı a. e' e eare a eve. , e t r gra f C ı a (Gra t 2021Y F A0909502 a 2020Y F A0907702) t e atı a. at ıra. S e e F ı atı f C ı a (Gra t 32270107 32071414 a 22075179) a t e atıra. S e e F ı a tı f S a g aı (21Z 1432100)

Conflict of Interest

T e a ıt r e are f.ı t f ı tere t

Data Availability Statement

T e ata t at ı , rt t e fı ı g f t ı t ı ' are avaı.a .e ı t e S ı , , rtı g , f r atı f t ı artı e

Keywords: Biomolecular Condensate · Membraneless Compartment · Quantum Dots · Spider Dragline Silk Protein · Synthetic Biology

[1] A fr e.e a ' A ı ı t , Me ı tz M ar T arr *Nat. Nanotechnol.* **2018** *13* 278–288
[2] / Ta.a ı A ga A r w..ı M aa e We..er *Nano Lett.* **2001** *1* 207–211
[3] Weg er ı. e ra t *Chem. Soc. Rev.* **2015** *44* 4792–4834
[4] Y C ı S Y ee *Nat. Chem. Rev.* **2020** *4* 638–656
[5] Tıa W W ı T T Z ı C e W Wa g F A Z a g C g Y Gıa F ı ● Z ı G

[6] Y C T ar. C ee SY ee *Proc. Natl. Acad. Sci. USA* **2018** *115* 5944–5949
[7] a g t e er *ACS Synth. Biol.* **2021** *10* 3475–3488
[8] C f Y Xie Z Z ag Y Y a g W a g Z X Xie C e f S e *Adv. Funct. Mater.* **2009** *19* 2359–2364
[9] S Stirze a M e er *Nat. Nanotechnol.* **2013** *8* 57–60
[10] f Xi g W T *Sci. China Chem.* **2020** *63* 448–453
[11] Tia Y Mi W W C e Z T T Z i Y Ma F A Z eg f ag Y Z f Y *ACS Nano* **2019** *13* 5841–5851
[12] S f a g z v M g A M a W C e *Angew. Chem. Int. Ed.* **2008** *47* 5186–5189 *Angew. Chem.* **2008** *120* 5264–5267
[13] T ar. SY ee S e T S Se *Angew. Chem. Int. Ed.* **2010** *49* 7019–7024 *Angew. Chem.* **2010** *122* 7173–7178
[14] S A. ert A G a fe ter T Mittag *Cell* **2019** *176* 419–434
[15] S F a a f, ee A A a M e *Nat. Rev. Mol. Cell Biol.* **2017** *18* 285–298
[16] ra a M T Wa.. C ra gw e *Nat. Biotechnol.* **2019** *37* 1435–1445
[17] Saar A S M rg v i W Arter G ra er A A ee T w e *Proc. Natl. Acad. Sci. USA* **2021** *118* e2019053118
[18] Y S C ra gw e *Science* **2017** *357* eaaf4382
[19] S We Z G ia C F f F a M T C e SY ee X X Xia *Nat. Chem. Biol.* **2020** *16* 1143–1148
[20] S S ter f ee art a arat C a e M Ca. we.. G er ez f a age M C G A f a er *Nat. Commun.* **2018** *9* 2985
[21] Me tz f T U e a G a f Matt *Nat. Mater.* **2005** *4* 435–446
[22] A M Wag er M e G , rive A e a *Acta Biomater.* **2019** *94* 44–63
[23] Y F e g f Wa g Z ag Y S g M Me g Mi f Y *Biomacromolecules* **2018** *19* 2432–2442
[24] W Y a g W G C a g Z a g *J. Mater. Chem. B* **2017** *5* 401–417
[25] , e a e e M a ae. q et C a f ae e C artie *Chem. Mater.* **2014** *26* 5383–5393
[26] M T f ee C S W ee *Colloids Surf. A* **2010** *359* 39–44
[27] Y arger C er A va er aart *Nat. Rev. Mater.* **2018** *3* 18008
[28] C F f Z G ia e g Y Z ag X X Xia *ACS Biomater. Sci. Eng.* **2021** *7* 3608–3617
[29] i F a C F f SY ee X X Xia Z G ia *Metab. Eng.* **2022** *70* 102–114
[30] M ia a a S e *Biochim. Biophys. Acta Proteins Proteomics* **2010** *1804* 1405–1412
[31] f re ter St e v f Mie e S M We C f r ar t Z ag G St y A f e *Environ. Sci. Technol.* **2009** *43* 2589–2594
[32] Ci Wa g f Wa g Y a g M Z a *J. Hazard. Mater.* **2021** *409* 124485
[33] , e.a e f e va f G at A a.. C t a arre Y ar *FEBS J.* **2010** *277* 5086–5096
[34] , r e Ae a a G A zia , t C eza a f za G zá.ez rav M érez *Front. Microbiol.* **2019** *10* 1587
[35] f g SY ee T S Se *Small* **2018** *14* 1803133

Ma r t re e ve Se te er 26 2022
A e te a r t e , t er 17 2022
er fre r e ve er 10 2022