Mimicking the chemistry of natural eumelanin synthesis: the KE sequence in polypeptides and in proteins allows for a specific control of nanosized functional polydopamine formation.

Camille Bergtold [1,2,], Daniel Hauser [3,*], Alain Chaumont[4], Salima Elyakhlifi[2], Mihaela Mateescu [1], Florent Meyer [1,2], Marie-Hélène Metz-Boutigue [1,2], Benoît Frisch [5], Pierre Schaaf [1,6], Dris Ihiawakrim [7], Ovidiu Ersen [7], Christophe A. Monnier[3], Alke Petri-Fink [3], Barbara Rothen-Rutishauser [3] & Vincent Ball [1,2,#]*

1: Université de Strasbourg, Faculté de Chirurgie Dentaire, 8 rue Sainte Elisabeth, 67000 Strasbourg, France.

2: Institut National de la Santé et de la Recherche Médicale, Unité Mixte de Recherche 1121, 11 rue Humann, 67085 Strasbourg Cedex, France.

3: Adolphe Merkle Institute, University of Fribourg, 4 Chemin des Verdiers, CH-1700 Fribourg, Switzerland.

4 : Faculté de Chimie, Chimie de la Matière Complexe, Centre National de la Recherche Scientifique, Unité Mixte de Recherche 7140, 4 rue Blaise Pascal, 67081, Strasbourg, France.

5: Université de Strasbourg, Faculté de Pharmacie, Laboratoire de Conception et application de molécules bioactives.

6: Centre National de la Recherche Scientifique, Institut Charles Sadron, Unité Propre 22, 23 rue du Loess, 67034 Strasbourg, France.

7: Centre National de la Recherche Scientifique, Institut de Physique et de Chimie des Matériaux, Unité Mixte de Recherche 7504, 23 rue du Loess, 67034, Strasbourg, Cedex 2, France.

email: vball@unistra.fr

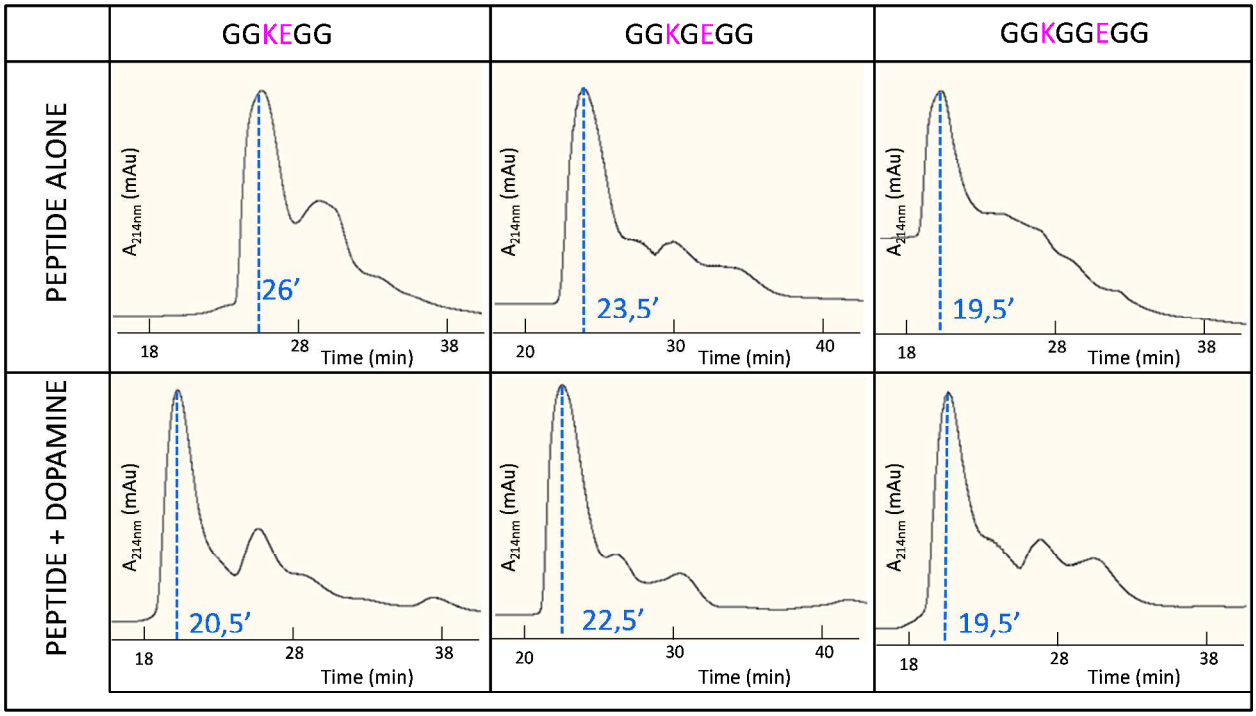

Figure SI 1: Size exclusion (SEC) chromatograms of the GGKEGG, GGKGEGG and GGKGGEGG peptides (first row) and of the same peptides with added dopamine in the presence of Tris buffer (pH = 8.5). The SEC were taken 16h after mixing dopamine with the considered peptides. The detection wavelength was set at 214 nm.

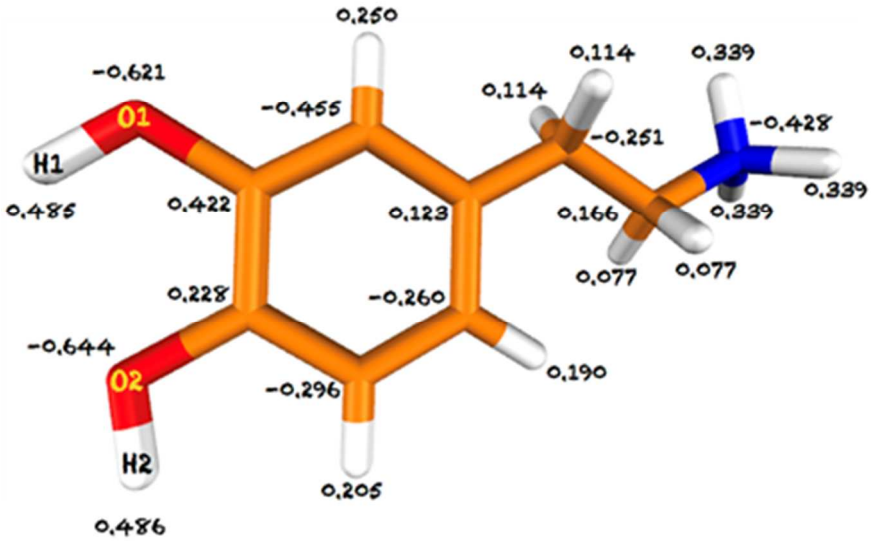

Figure SI 2: Schematic representation of the dopamine molecule and the RESP charges used during MD simulation.

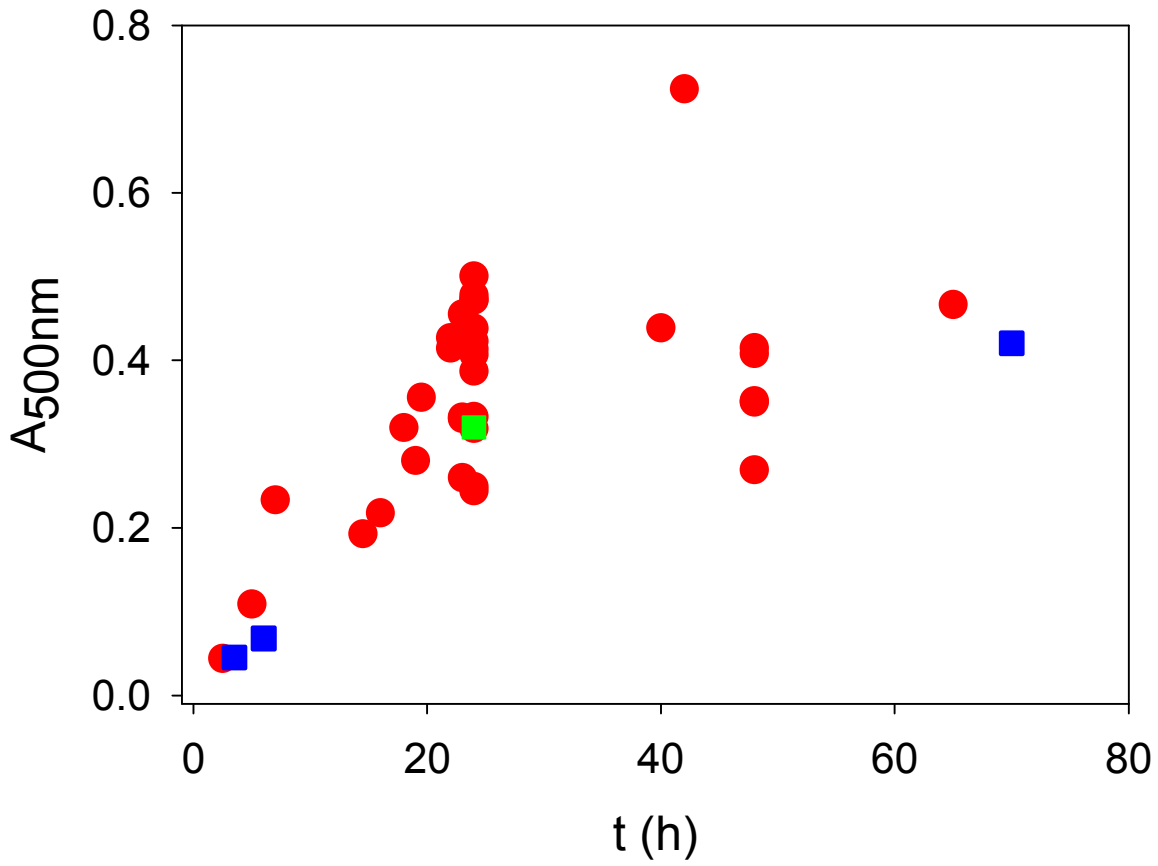

Figure SI 3: Absorbance of PDA films deposited on quartz slides recorded at λ=500 nm as a function of the oxidation time in the presence of dopamine at 2mg.mL^{-1} (Tris buffer 50 mM, pH = 8.5), in the absence of lysozyme (●) and in the presence of lysozyme at 0.5 (■) and 1 (■) mg.mL^{-1}.

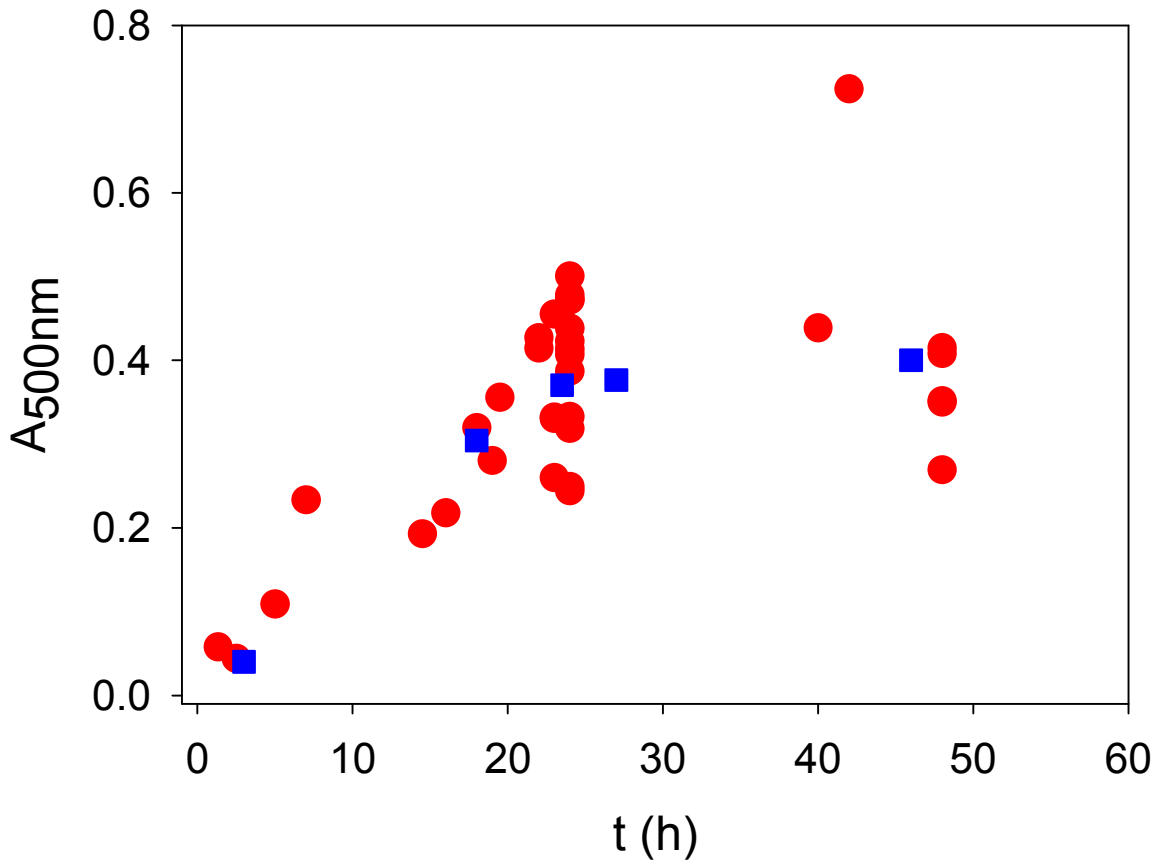

Figure SI 4: Absorbance of PDA films deposited on quartz slides recorded at λ=500 nm as a function of the oxidation time in the presence of dopamine at 2mg.mL^{-1} (Tris buffer 50 mM, pH = 8.5), in the absence of α lactalbumin (●) and in the presence of α lactalbumin at 1.0 mg.mL^{-1} (■).

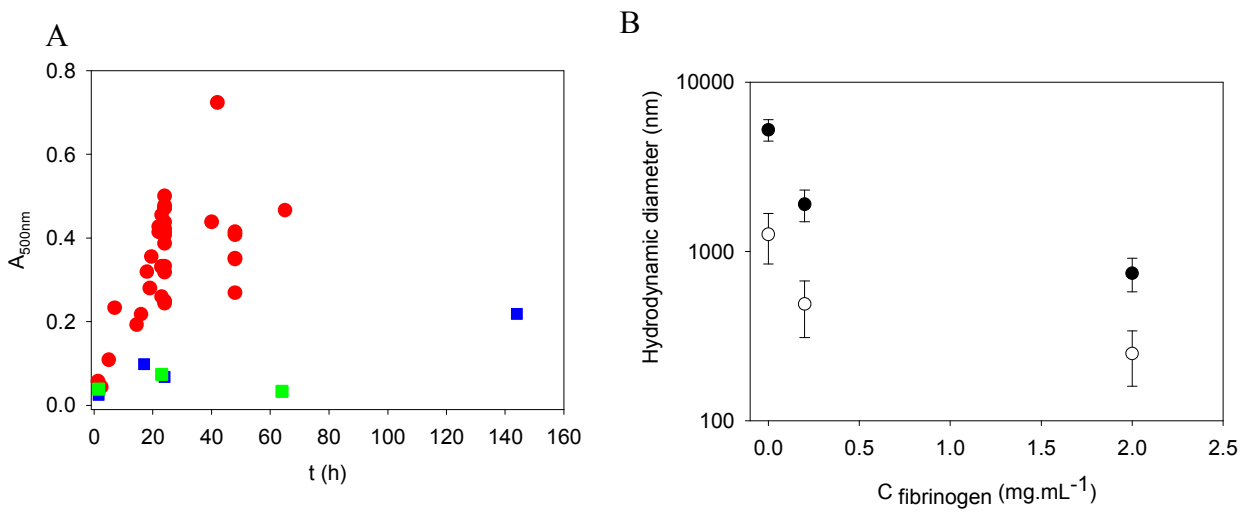

Figure SI 5: A : Absorbance of PDA films deposited on quartz slides recorded at λ=500 nm as a function of the oxidation time in the presence of dopamine at 2mg.mL^{-1} (Tris buffer 50 mM, pH = 8.5), in the absence of human fibrinogen (●) and in the presence of human fibrinogen at 0.2 mg.mL^{-1} (■) and at 2 mg.mL-1 (■).

B: Evolution of the hydrodynamic diameter of PDA@fibrinogen particles after 24 h of oxidation (2 mg.mL^{-1} dopamine in the presence of 50 mM Tris buffer at pH = 8.5) as a function of the protein concentration. The protein + PDA mixture was dialyzed before the light scattering experiments. (○) and (●) correspond to the smaller and larger particles with relative fractions of about 60 and 40 % (in number of particles)

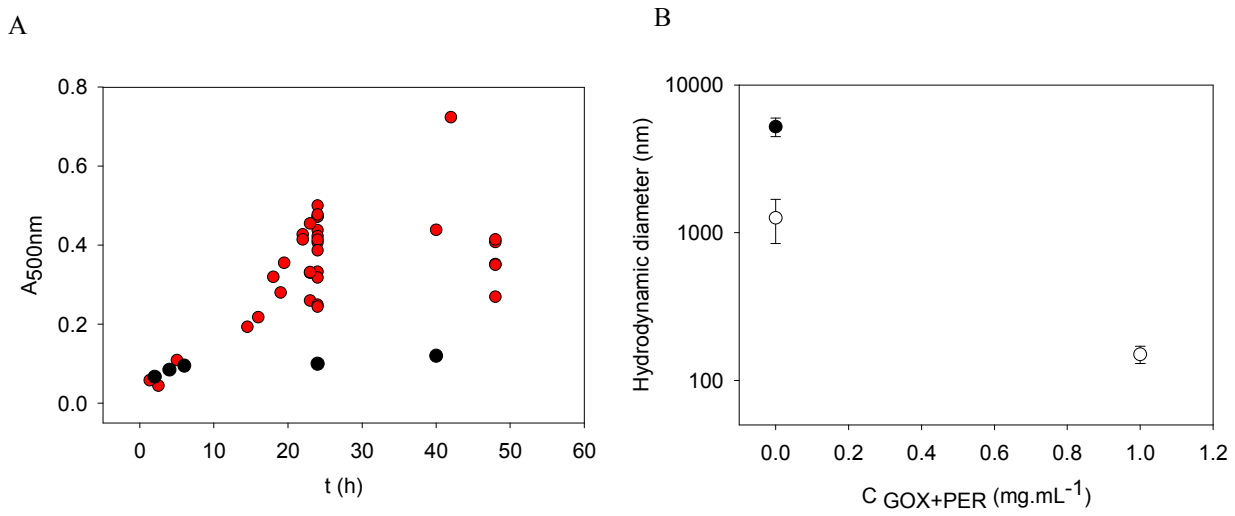

Figure SI 6: A : Absorbance of PDA films deposited on quartz slides recorded at λ=500 nm as a function of the oxidation time in the presence of dopamine at 2mg.mL^{-1} (Tris buffer 50 mM, pH = 8.5), in the absence of glucose oxidase + peroxidase (●) and in the presence of a mixture of both proteins at 0.5 mg.mL^{-1} each (●).

B: Evolution of the hydrodynamic diameter of PDA@GOX+POX particles after 24 h of oxidation (2 mg.mL^{-1} dopamine in the presence of 50 mM Tris buffer at pH = 8.5) as a function of the protein concentration. The protein + PDA mixture was dialyzed before the light scattering experiments. (○) and (●) correspond to the smaller and larger particles with relative fractions of about 60 and 40 % (in number of particles) in the absence of proteins and to 100 % in the presence of GOX (0.5 mg.mL^{-1}) and POX (0.5 mg.mL^{-1}).

Human Serum Albumin

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         10         20         30         40         50
QFPTDYDEGQ DDRPKVGLGA RGHRPYDKKK EEAPSLRPVP PPISGGGYRA
         60         70         80         90        100
RPATATVGQK KVERKPPDAD GCLHADPDLG VLCPTGCKLQ DTLVRQERPI
        110        120        130        140        150
RKSIEDLRNT VDSVSRTSSS TFQYITLLKN MWKGRQNQVQ DNENVVNEYS
        160        170        180        190        200
SHLEKHQLYI DETVKNNIPT KLRVLRSILE NLRSKIQKLE SDVSTQMEYC
        210        220        230        240        250
RTPCTVTCNI PVVSGKECEK IIRNEGETSE MYLIQPEDSS KPYRVYCDMK
        260        270        280        290        300
TEKGGWTVIQ NRQDGSVDFG RKWDPYKQGF GNIATNAEGK KYCGVPGEYW
        310        320        330        340        350
LGNDRISQLT NMGPTKLLIE MEDWKGDKVT ALYEGFTVQN EANKYQLSVS
        360        370        380        390        400
KYKGTAGNAL IEGASQLVGE NRTMTIHNSM FFSTYDRDND GWKTTDPRKQ
        410        420        430        440        450
CSKEDGGGWW YNRCHAANPN GRYYWGGAYT WDMAKHGTDD GVVWMNWQGS
        460
WYSMKKMSMK IRPYFPEQ
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Glucose oxidase (Aspergillus Niger)

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         10         20         30         40         50
MQTLLVSSLV VSLAAALPHY IRSNGIEASL LTDPKDVSGR TVDYIIAGGG
         60         70         80         90        100
LTGLTTAARL TENPNISVLV IESGSYESDR GPIIEDLNAY GDIFGSSVDH
        110        120        130        140        150
AYETVELATN NQTALIRSGN GLGGSTLVNG GTWTRPHKAQ VDSWETVFGN
        160        170        180        190        200
EGWNWDNVAA YSLQAERARA PNAKQIAAGH YFNASCHGVN GTVHAGPRDT
        210        220        230        240        250
GDDYSPIVKA LMSAVEDRGV PTKKDFGCGD PHGVSMFPNT LHEDQVRSDA
        260        270        280        290        300
AREWLLPNYQ RPNLQVLTGQ YVGKVLLSQN GTTPRAVGVE FGTHKGNTHN
        310        320        330        340        350
VYAKHEVLLA AGSAVSPTIL EYSGIGMKSI LEPLGIDTVV DLPVGLNLQD
        360        370        380        390        400
QTTATVRSRI TSAGAGQGQA AWFATFNETF GDYSEKAHEL LNTKLEQWAE
        410        420        430        440        450
EAVARGGFHN TTALLIQYEN YRDWIVNHNV AYSELFLDTA GVASFDVWDL
        460        470        480        490        500
LPFTRGYVHI LDKDPYLHHF AYDPQYFLNE LDLLGQAAAT QLARNISNSG
        510        520        530        540        550
AMQTYFAGET IPGDNLAYDA DLSAWTEYIP YHFRPNYHGV GTCSMMPKEM
        560        570        580        590        600
GGVVDNAARV YGVQGLRVID GSIPPTQMSS HVMTVFYAMA LKISDAILED

YASMQ
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Horseradish peroxidase

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         10         20         30         40         50
MAMSYSIRVL TFLMLISLMA VTLNLLSTAE AKKPRRDVPI VKGLSWNFYQ
         60         70         80         90        100
RACPKVEKII KKELKKVFKR DIGLAAAILR IHFHDCFVQG CEASVLLAGS
        110        120        130        140        150
ASGPGEQSSI PNLTLRQQAF VVINNLRALV QKQCGQVVSC SDILALAARD
        160        170        180        190        200
SIVLSGGPDY AVPLGRRDSL AFATPETTLA NLPPPFANAS QLISDFNDRN
        210        220        230        240        250
LNITDLVALS GGHTIGIAHC PSFTDRLYPN QDPTMNKSFA NSLKRTCPTA
        260        270        280        290        300
NSSNTQVNDI RSPDVFDNKY YVDLMNRQGL FTSDQDLFVD KRTRGIVESF
        310        320        330        340        350
AIDQNLFFDH FTVAMIKMGQ MSVLTGTQGE IRSNCSARNT ASFISVLEEG

IVEEALSMI
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Hemoglobin α chain

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         10          20          30          40          50
MVLSPADKTN VKAAWGKVGA HAGEYGAEAL ERMFLSFPTT KTYFPHFDLS
         60          70          80          90         100
HGSAQVKGHG KKVADALTNA VAHVDDMPNA LSALSDLHAH KLRVDPVNFK
        110         120         130         140
LLSHCLLVTL AAHLPAEFTP AVHASLDKFL ASVSTVLTSK YR
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Figure SI 7: Amino acid sequences of the proteins used to control the oxidation of dopamine and its assembly in PDA. The KE sequence has been highlighted in red when present.

The sequences are extracted from UniprotKB.

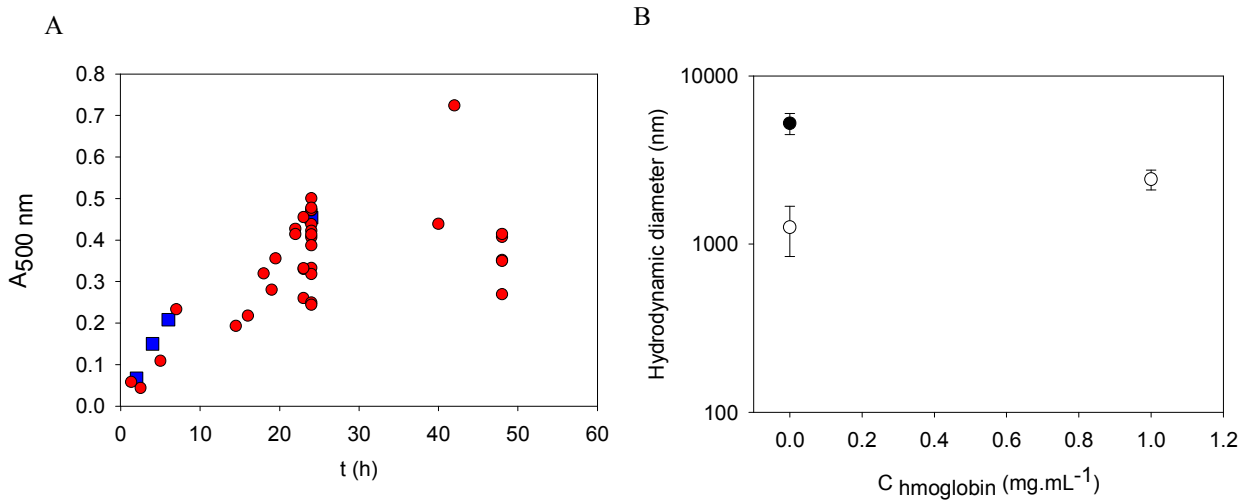

Figure SI 8 : A : Absorbance of PDA films deposited on quartz slides recorded at λ=500 nm as a function of the oxidation time in the presence of dopamine at 2mg.mL^{-1} (Tris buffer 50 mM, pH = 8.5), in the absence of human hemogmlobin (●) and in the presence of human hemoglobin at 0.2 mg.mL^{-1} (■).

B: Evolution of the hydrodynamic diameter of PDA@hemoglobin particles after 24 h of oxidation (2 mg.mL^{-1} dopamine in the presence of 50 mM Tris buffer at pH = 8.5) as a function of the protein concentration. The protein + PDA mixture was dialyzed before the light scattering

experiments. (○) and (●) correspond to the smaller and larger particles with relative fractions of about 60 and 40 % (in number of particles)