Supplementary Material S1. Wang *et al.* kinetic scheme

Pellets are classified in terms of length 1, 2, …, 6. Breakage is first order with respect to the number of mother pellets n_i and gives two daughter pellets, of length j and k where $i = j + k$, with combinations of outcomes shown in Figure S1.

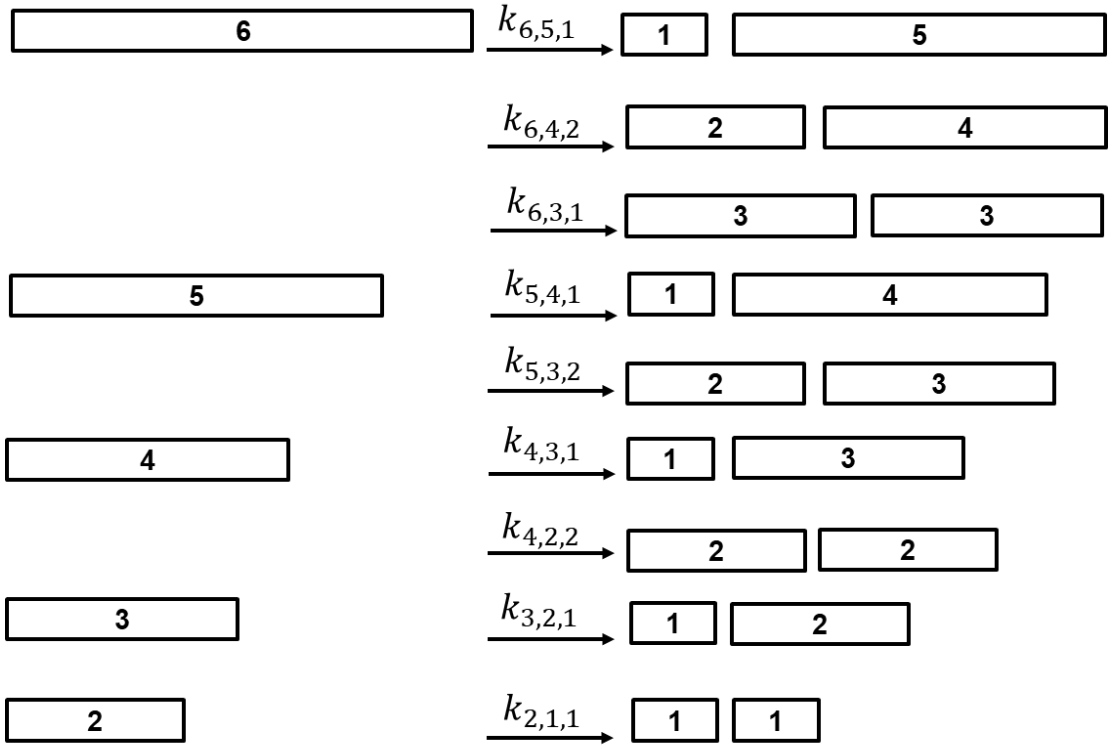


Figure S1 Schematic of pellet length evolution. $k_{i,j,i-j}$ is the first order breakage constant, where *i* refers to the length of the mother pellet and *j* the length of the longer daughter pellet.

The one-dimensional population balance equations are

$$\frac{dn_6}{d\tau} = -n_6\left(k_{6,5,1} + k_{6,4,2} + k_{6,3,3}\right) \quad \text{(S1)}$$

$$\frac{dn_5}{d\tau} = -n_5\left(k_{5,4,1} + k_{5,3,2}\right) + n_6 k_{6,5,1} \quad \text{(S2)}$$

$$\frac{dn_4}{d\tau} = -n_4\left(k_{4,3,1} + k_{4,2,2}\right) + n_6 k_{6,4,2} + n_5 k_{5,4,1} \quad \text{(S3)}$$

$$\frac{dn_3}{d\tau} = -n_3 k_{3,2,1} + 2n_6 k_{6,3,3} + n_5 k_{5,3,2} + n_4 k_{4,3,1} \quad \text{(S4)}$$

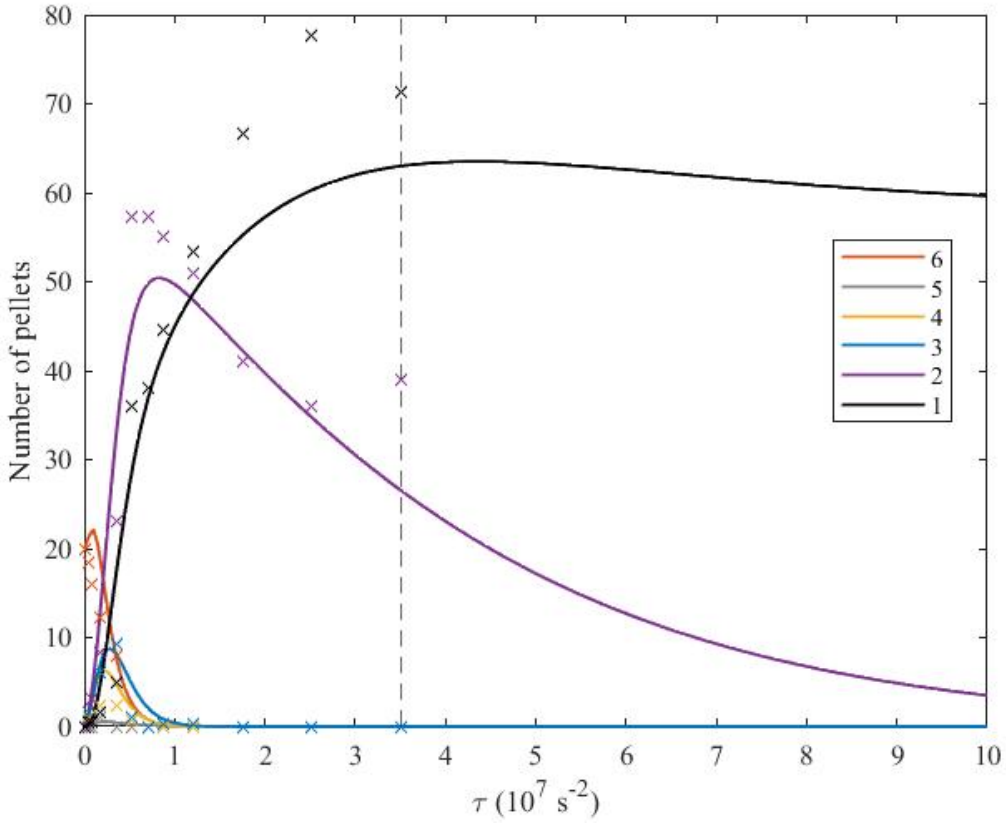
$$\frac{dn_2}{d\tau} = -n_2 k_{2,1,1} + n_6 k_{6,4,2} + n_5 k_{5,3,2} + 2n_4 k_{4,2,2} + n_3 k_{3,2,1} \quad \text{(S5)}$$

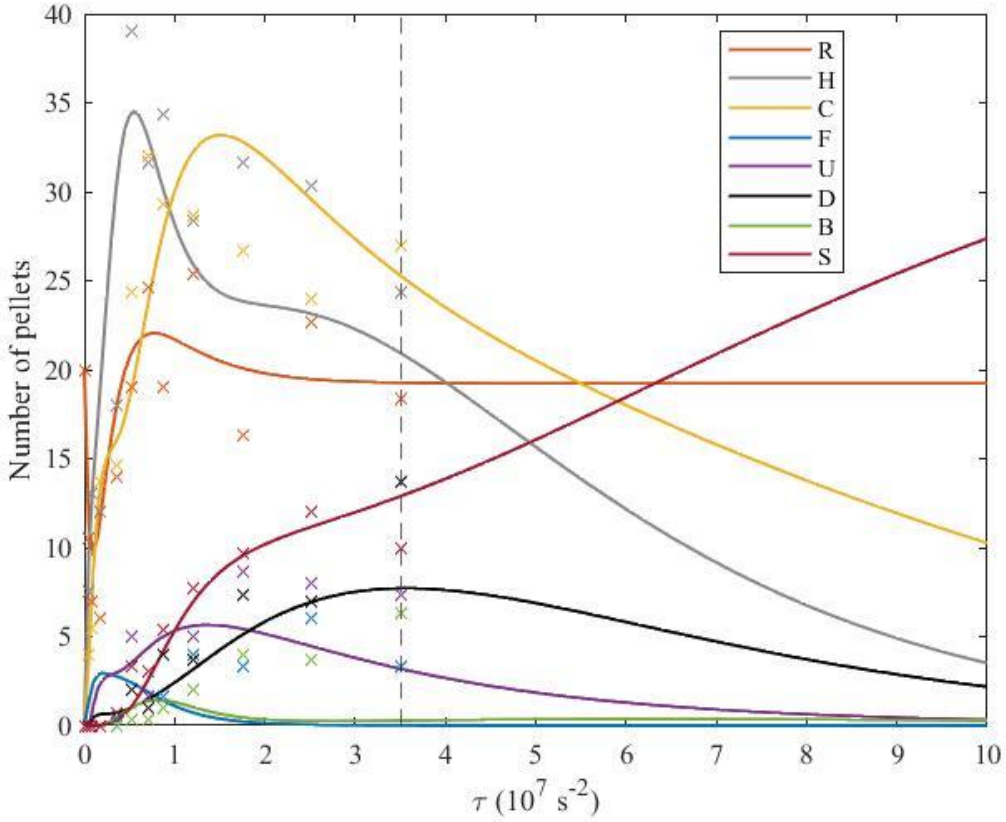
$$\frac{dn_1}{d\tau} = n_6 k_{6,5,1} + n_5 k_{5,4,1} + n_4 k_{4,3,1} + n_3 k_{3,2,1} + 2n_2 k_{2,1,1} \quad \text{(S6)}$$

where τ is scaled time.

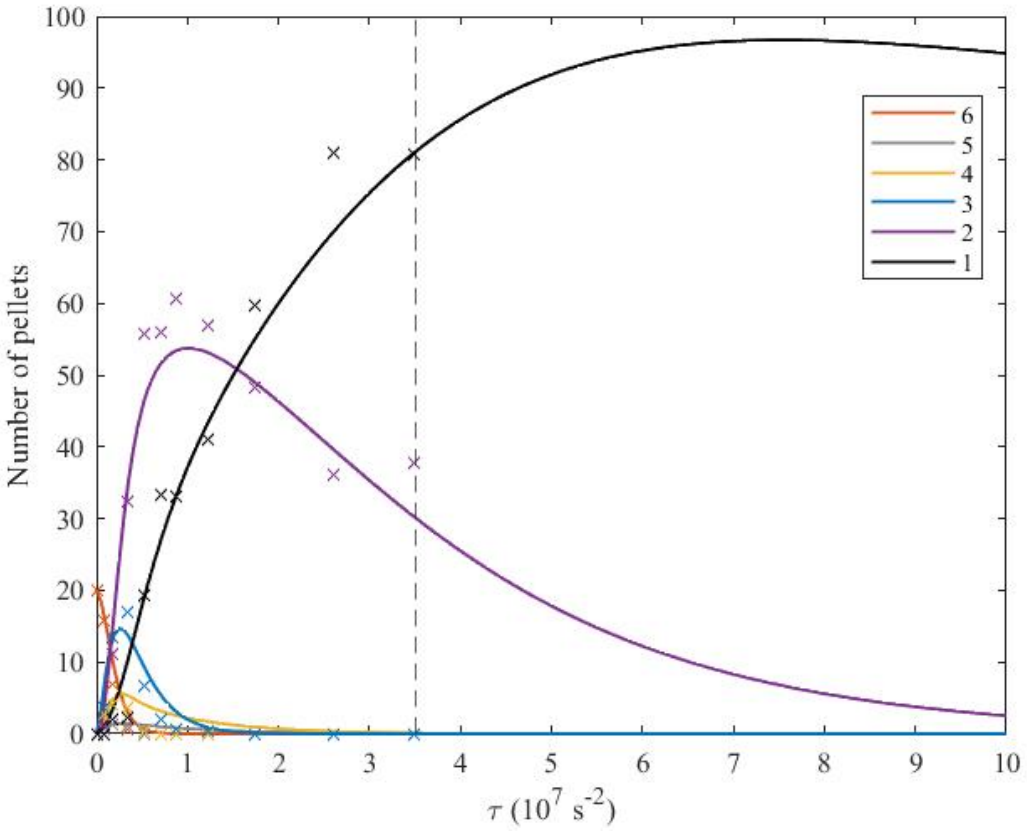
Supplementary Material S2 Pellet Evolution for other cases

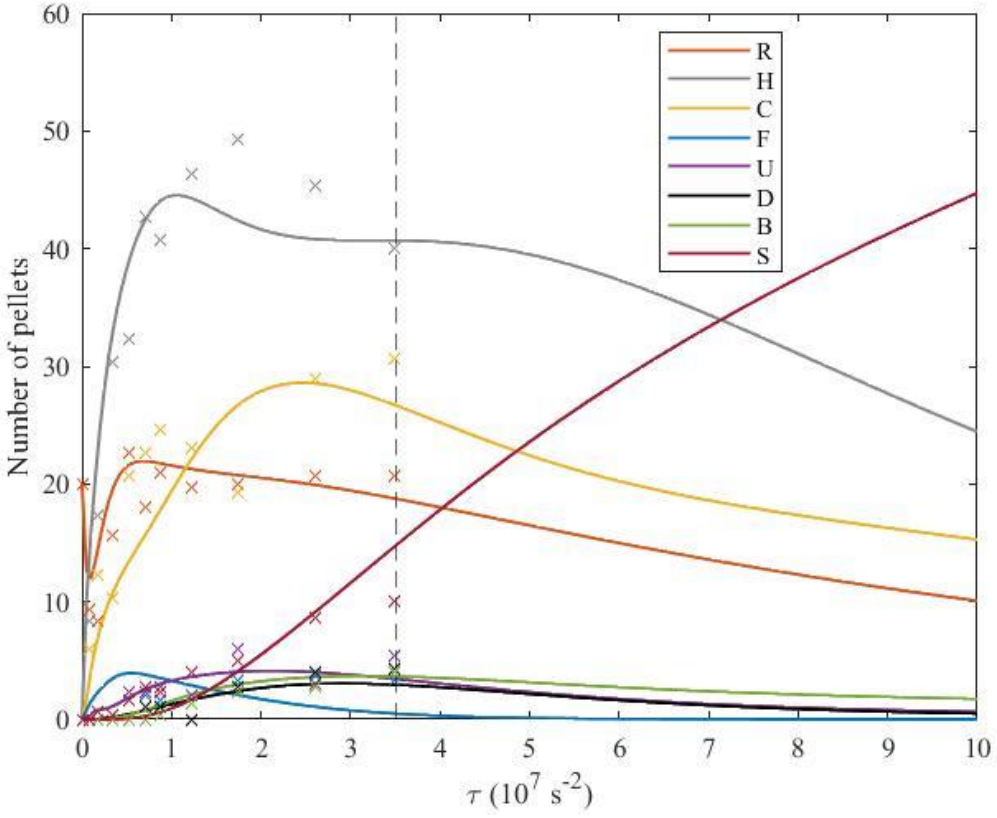
Case I



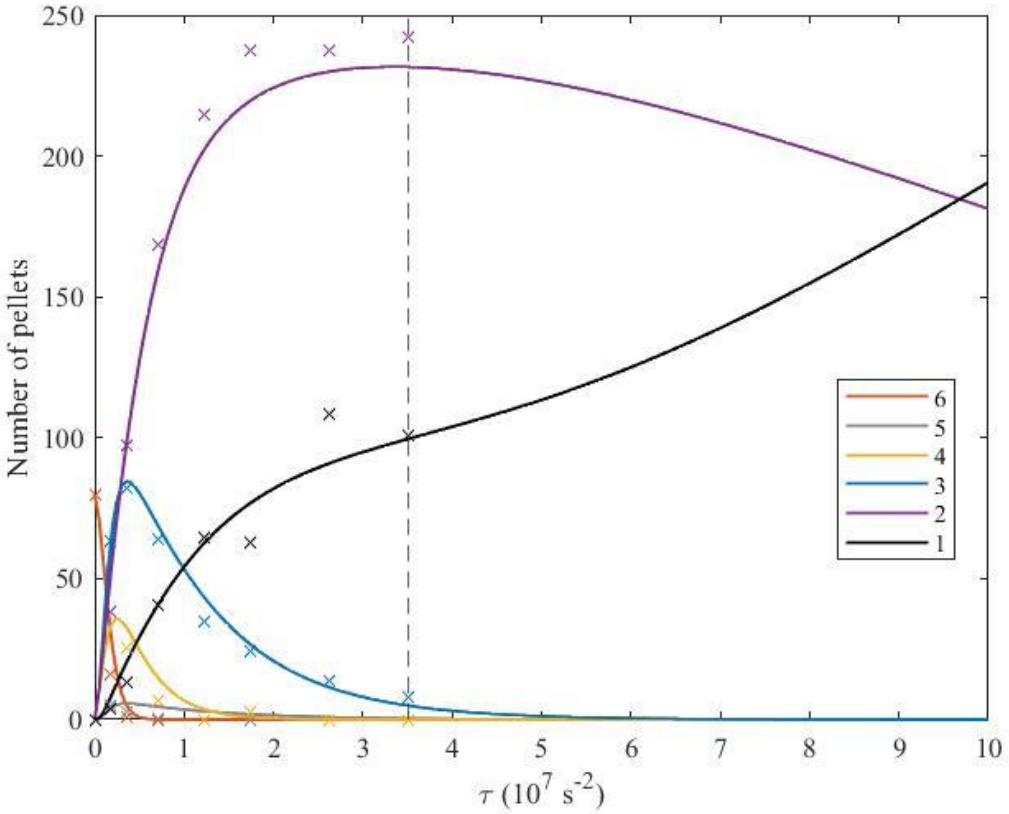


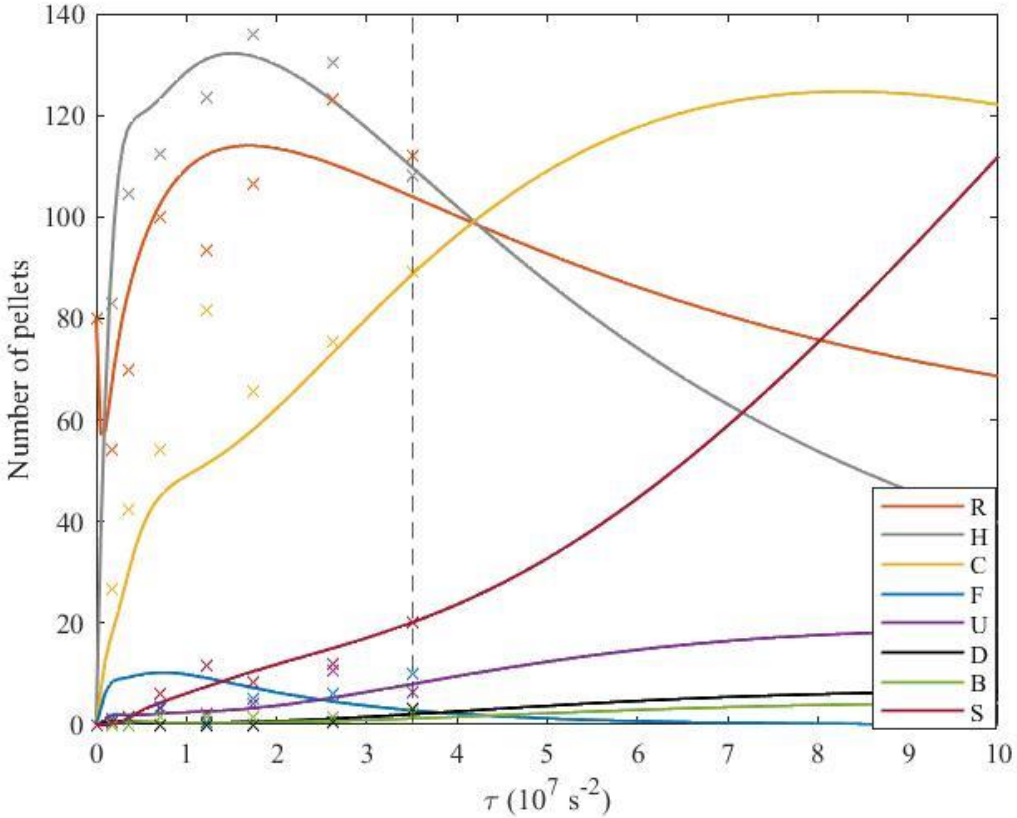
Case II


Number of pellets
τ (10^7 s^{-2})
6
5
4
3
2
1

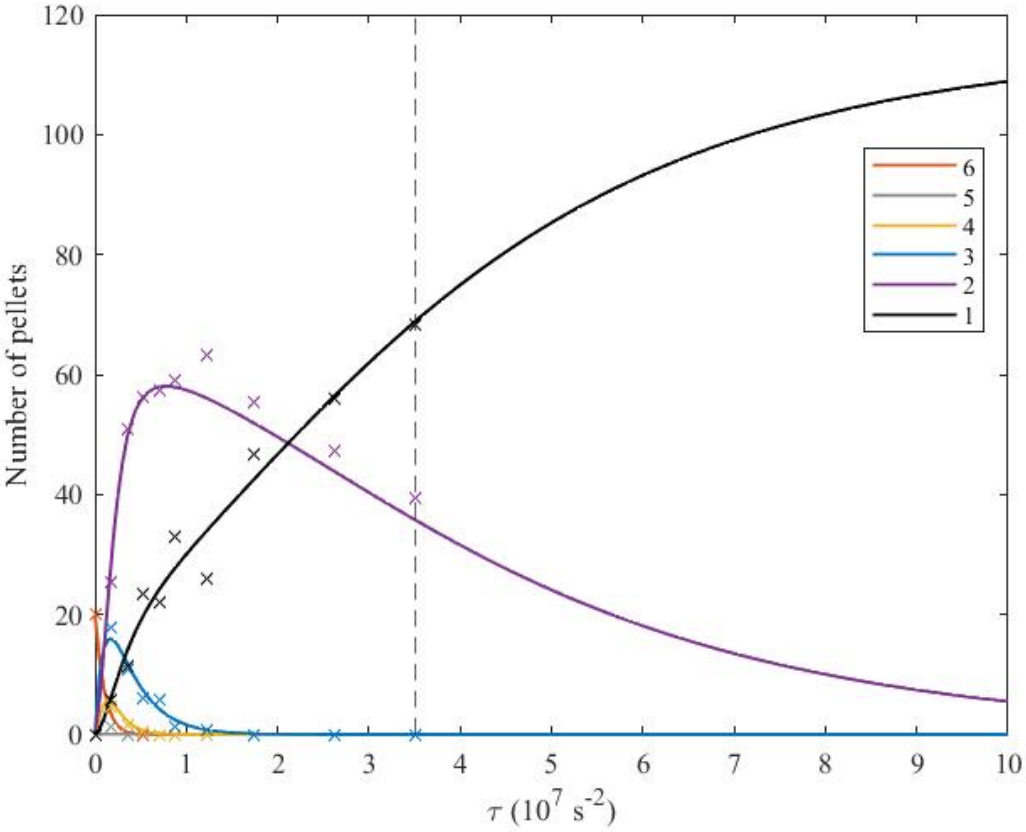

Number of pellets
τ (10^7 s^{-2})
R
H
C
F
U
D
B
S

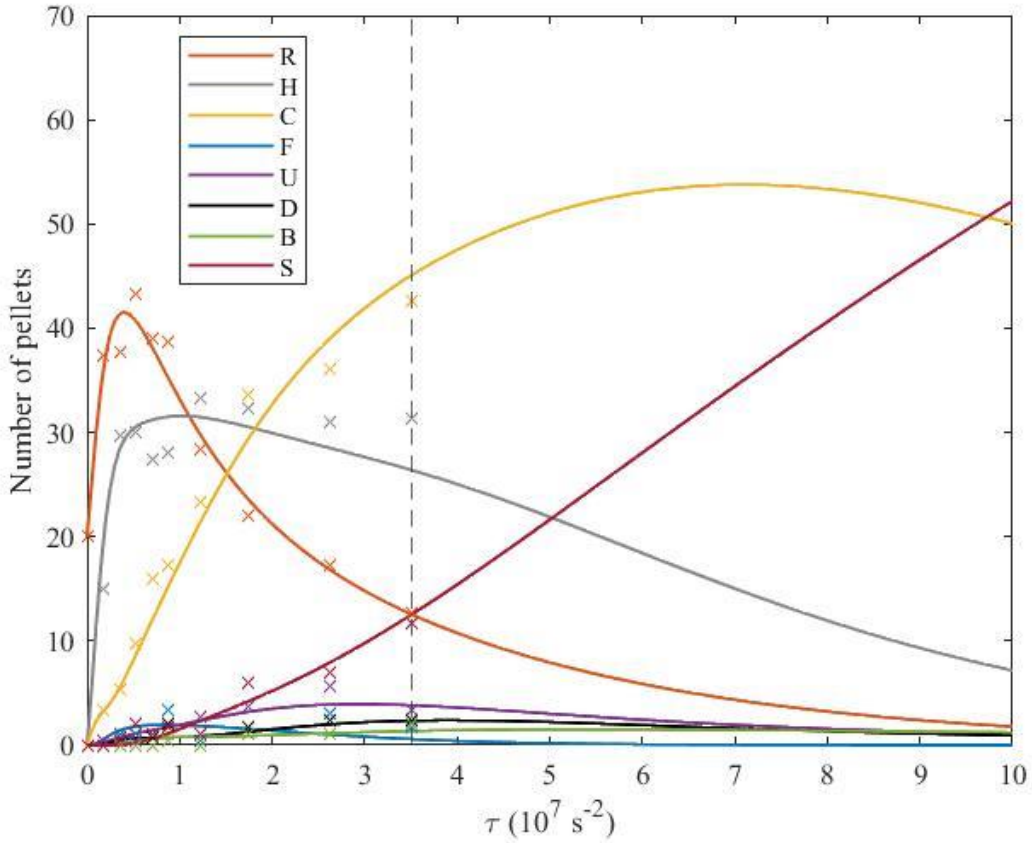
Case IIIb


Number of pellets
τ (10^7 s^-2)
6
5
4
3
2
1


Number of pellets
τ (10^7 s^-2)
R
H
C
F
U
D
B
S

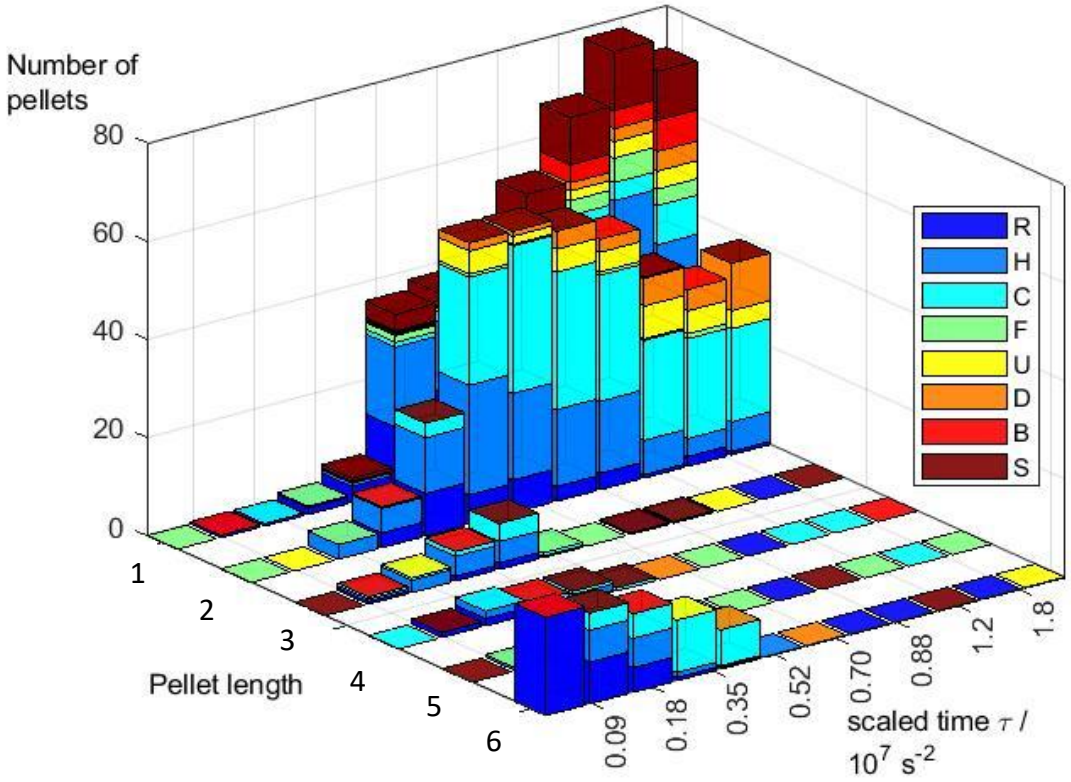
Case IV


Number of pellets
τ (10^7 s^{-2})
6
5
4
3
2
1

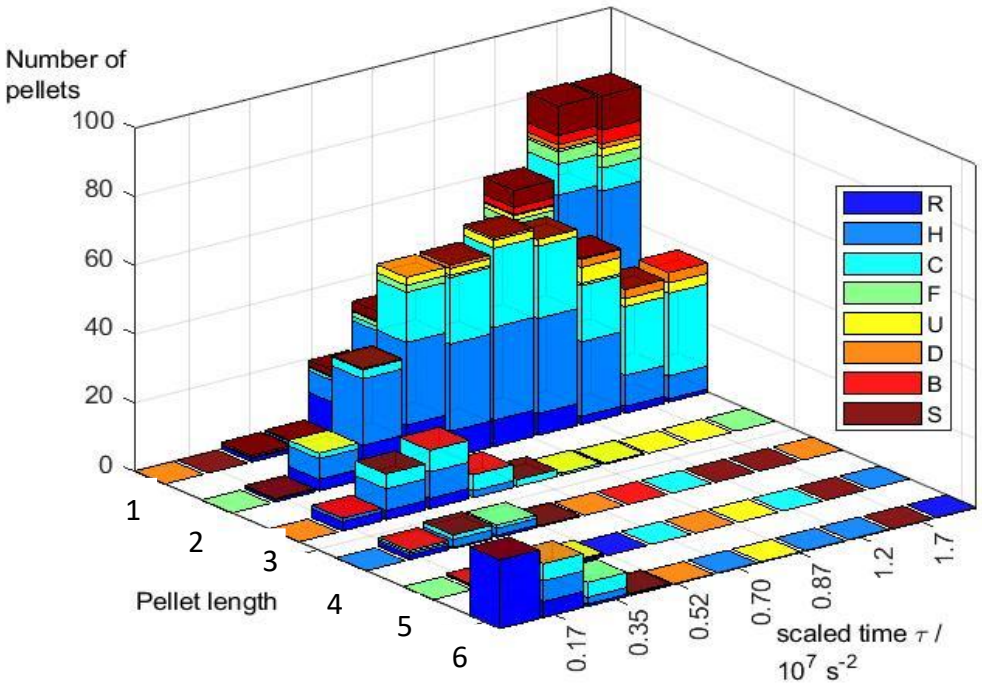

Number of pellets
τ (10^7 s^{-2})
R
H
C
F
U
D
B
S

Supplementary Material S3. Pellet Evolution for other Cases

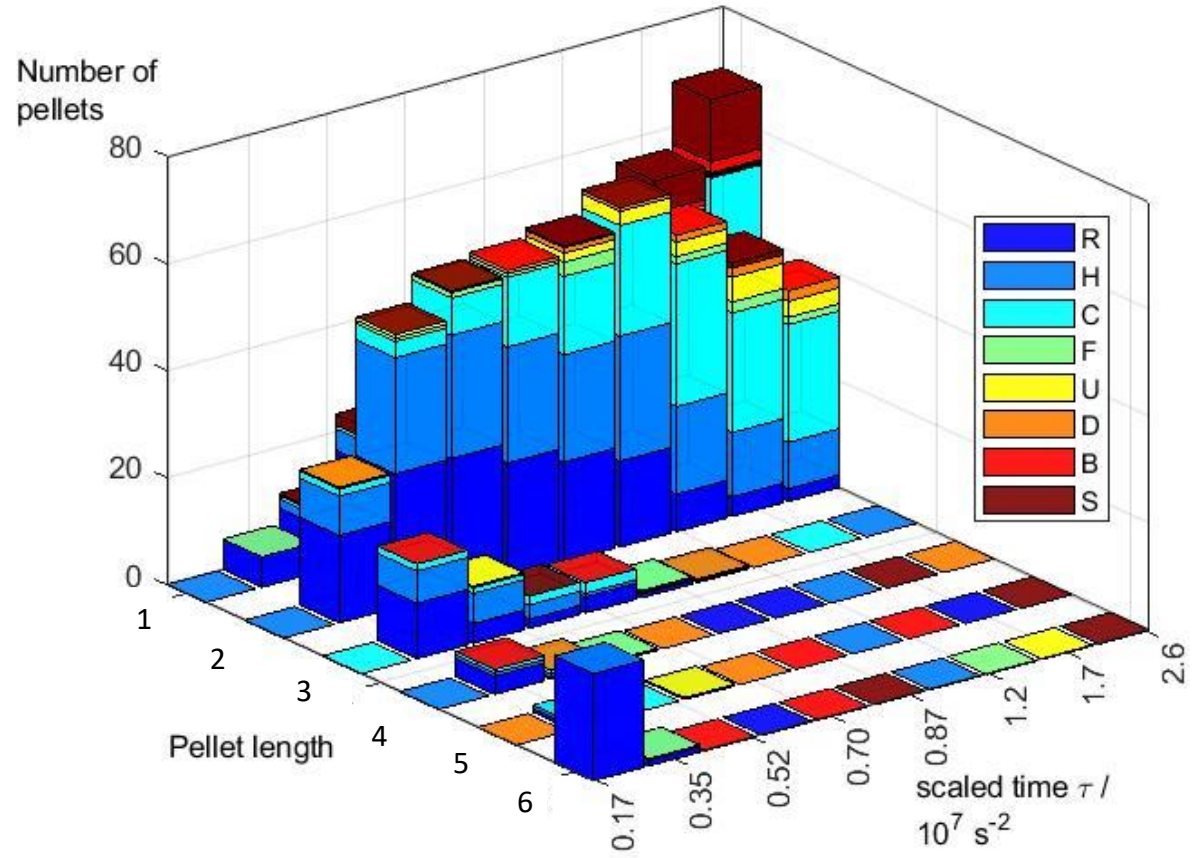
(a) Case I



(b) Case II



(c) Case IV


Number of pellets
80
60
40
20
0
1
2
3
4
5
6
Pellet length
0.17
0.35
0.52
0.70
0.87
1.2
1.7
2.6
scaled time τ / 10^7 s^-2
R
H
C
F
U
D
B
S

Supplementary material S4. Matlab code for 2-D Population Balance Model in Appendix 1

```
kmatrix(1,1) = -(k_6R_6H + k_6R_5R_1R + k_6R_4R_2R + k_6R_3R_3R);

kmatrix(2,1) = k_6R_5R_1R;
kmatrix(2,7) = k_6H_5R_1H;
kmatrix(2,19) = k_6F_5R_1F;
kmatrix(2,2) = -(k_5R_5H + k_5R_4R_1R + k_5R_3R_2R);

kmatrix(3,3) = -(k_4R_4H + k_4R_3R_1R + k_4R_2R_2R);
kmatrix(3,1) = k_6R_4R_2R;
kmatrix(3,2) = k_5R_4R_1R;
kmatrix(3,7) = k_6H_4R_2H;
kmatrix(3,8) = k_5H_4R_1H;
kmatrix(3,19) = k_6F_4R_2F;
kmatrix(3,20) = k_5F_4R_1F;

kmatrix(4,4) = -(k_3R_3H + k_3R_2R_1R);
kmatrix(4,1) = 2*k_6R_3R_3R;
kmatrix(4,2) = k_5R_3R_2R;
kmatrix(4,3) = k_4R_3R_1R;
kmatrix(4,7) = k_6H_3R_3H;
kmatrix(4,8) = k_5H_3R_2H;
kmatrix(4,9) = k_4H_3R_1H;
kmatrix(4,19) = k_6F_3R_3F;
kmatrix(4,20) = k_5F_3R_2F;
kmatrix(4,21) = k_4F_3R_1F;

kmatrix(5,5) = -(k_2R_2H + k_2R_1R_1R);
kmatrix(5,1) = k_6R_4R_2R;
kmatrix(5,2) = k_5R_3R_2R;
kmatrix(5,3) = 2*k_4R_2R_2R;
kmatrix(5,4) = k_3R_2R_1R;
kmatrix(5,7) = k_6H_4R_2H;
kmatrix(5,8) = k_5H_3R_2H;
kmatrix(5,9) = k_4H_2R_2H;
kmatrix(5,10) = k_3H_2R_1H;
kmatrix(6,19) = k_6F_4R_2F;
kmatrix(5,20) = k_5F_3R_2F;
kmatrix(5,21) = k_4F_2R_2F;
kmatrix(5,22) = k_3F_2R_1F;

kmatrix(6,6) = -(k_1R_1H);
kmatrix(6,1) = k_6R_5R_1R;
kmatrix(6,2) = k_5R_4R_1R;
kmatrix(6,3) = k_4R_3R_1R;
kmatrix(6,4) = k_3R_2R_1R;
kmatrix(6,5) = 2*k_2R_1R_1R;
kmatrix(6,7) = k_6H_5R_1H;
kmatrix(6,8) = k_5H_4R_1H;
kmatrix(6,9) = k_4H_3R_1H;
kmatrix(6,10) = k_3H_2R_1H;
```

```
kmatrix(6,11) = k_2H_1R_1H;
kmatrix(6,19) = k_6F_5R_1F;
kmatrix(6,20) = k_5F_4R_1F;
kmatrix(6,21) = k_4F_3R_1F;
kmatrix(6,22) = k_3F_2R_1F;
kmatrix(6,23) = k_2F_1R_1F;

kmatrix(7,7) = -((2*k_6H_5R_1H) + (2*k_6H_4R_2H) + k_6H_3R_3H + k_6H_6F);
kmatrix(7,1) = k_6R_6H;

kmatrix(8,8) = -(2*k_5H_4R_1H + 2*k_5H_3R_2H + k_5H_5C + k_5H_5F);
kmatrix(8,2) = k_5R_5H;
kmatrix(8,7) = k_6H_5R_1H;
kmatrix(8,13) = k_6C_5H_1H;
kmatrix(8,25) = k_6U_5H_1F;

kmatrix(9,9) = -(2*k_4H_3R_1H + k_4H_2R_2H + k_4H_4X + k_4H_4C);
kmatrix(9,3) = k_4R_4H;
kmatrix(9,7) = k_6H_4R_2H;
kmatrix(9,8) = k_5H_4R_1H;
kmatrix(9,13) = k_6C_4H_2H;
kmatrix(9,14) = k_5C_4H_1H;
kmatrix(9,25) = k_6U_4H_2F;
kmatrix(9,26) = k_5U_4H_1F;

kmatrix(10,10) = -(2*k_3H_2R_1H + k_3H_3C + k_3H_3F);
kmatrix(10,4) = k_3R_3H;
kmatrix(10,7) = k_6H_3R_3H;
kmatrix(10,8) = k_5H_3R_2H;
kmatrix(10,9) = k_4H_3R_1H;
kmatrix(10,13) = 2*k_6C_3H_3H;
kmatrix(10,14) = k_5C_3H_2H;
kmatrix(10,25) = k_6U_3H_3F;
kmatrix(10,26) = k_5U_3H_2F;

kmatrix(11,11) = -(k_2H_1R_1H + k_2H_2C + k_2H_2F);
kmatrix(11,5) = k_2R_2H;
kmatrix(11,7) = k_6H_4R_2H;
kmatrix(11,8) = k_5H_3R_2H;
kmatrix(11,9) = k_4H_2R_2H;
kmatrix(11,10) = k_3H_2R_1H;
kmatrix(11,13) = k_6C_4H_2H;
kmatrix(11,14) = k_5C_3H_2H;
kmatrix(11,15) = 2*k_4C_2H_2H;
kmatrix(11,16) = k_3C_2H_1H;
kmatrix(11,25) = k_6U_4H_2F;
kmatrix(11,26) = k_5U_3H_2F;
kmatrix(11,27) = k_4U_2H_2F;
kmatrix(11,28) = k_3U_2H_1F;

kmatrix(12,12) = -(k_1H_1C + k_1H_1F);
```

```
kmatrix(12,6) = k_1R_1H;
kmatrix(12,7) = k_6H_5R_1H;
kmatrix(12,8) = k_5H_4R_1H;
kmatrix(12,9) = k_4H_3R_1H;
kmatrix(12,10) = k_3H_2R_1H;
kmatrix(12,11) = k_2H_1R_1H;
kmatrix(12,13) = k_6C_5H_1H;
kmatrix(12,14) = k_5C_4H_1H;
kmatrix(12,15) = k_4C_3H_1H;
kmatrix(12,16) = k_3C_2H_1H;
kmatrix(12,17) = 2*k_2C_1H_1H;
kmatrix(12,25) = k_6U_5H_1F;
kmatrix(12,26) = k_5U_4H_1F;
kmatrix(12,27) = k_4U_3H_1F;
kmatrix(12,28) = k_3U_2H_1F;
kmatrix(12,29) = k_2U_1H_1F;

kmatrix(13,13) = -(k_6C_5H_1H + k_6C_4H_2H + k_6C_3H_3H + k_6C_6U);
kmatrix(13,7) = k_6H_6C;

kmatrix(14,14) = -(k_5C_4H_1H + k_5C_3H_2H + k_5C_5U);
kmatrix(14,8) = k_5H_5C;

kmatrix(15,15) = -(k_4C_3H_1H + k_4C_2H_2H + k_4C_4U);
kmatrix(15,9) = k_4H_4C;

kmatrix(16,16) = -(k_3C_2H_1H + k_3C_3U);
kmatrix(16,10) = k_3H_3C;

kmatrix(17,17) = -(k_2C_1H_1H + k_2C_2U);
kmatrix(17,11) = k_2H_2C;

kmatrix(18,18) = -k_1C_1U;
kmatrix(18,12) = k_1H_1C;

kmatrix(19,19) = -(2*k_6F_5R_1F + 2*k_6F_4R_2F + k_6F_3R_3F + k_6F_6U);
kmatrix(19,7) = k_6H_6F;

kmatrix(20,20) = -(2*k_5F_4R_1F + 2*k_5F_3R_2F + k_5F_5U);
kmatrix(20,8) = k_5H_5F;
kmatrix(20,19) = k_6F_5R_1F;

kmatrix(21,21) = -(2*k_4F_3R_1F + k_4F_2R_2F + k_4F_4U);
kmatrix(21,9) = k_4H_4F;
kmatrix(21,19) = k_6F_4R_2F;
kmatrix(21,20) = k_5F_4R_1F;

kmatrix(22,22) = -(2*k_3F_2R_1F + k_3F_3U);
kmatrix(22,10) = k_3H_3F;
kmatrix(22,19) = k_6F_3R_3F;
kmatrix(22,20) = k_5F_3R_2F;
```

```
kmatrix(22,21) = k_4F_3R_1F;

kmatrix(23,23) = -(k_2F_1R_1F + k_2F_2U);
kmatrix(23,11) = k_2H_2F;
kmatrix(23,19) = k_6F_4R_2F;
kmatrix(23,20) = k_5F_3R_2F;
kmatrix(23,21) = k_4F_2R_2F;
kmatrix(23,22) = k_3F_2R_1F;

kmatrix(24,24) = -(k_1F_1U);
kmatrix(24,11) = k_2H_2F;
kmatrix(24,19) = k_6F_5R_1F;
kmatrix(24,20) = k_5F_4R_1F;
kmatrix(24,21) = k_4F_3R_1F;
kmatrix(24,22) = k_3F_2R_1F;
kmatrix(24,23) = k_2F_1R_1F;

kmatrix(25,25) = -(2*k_6U_5H_1F + 2*k_6U_4H_2F + k_6U_3H_3F + k_6U_6D);
kmatrix(25,13) = k_6C_6U;
kmatrix(25,19) = k_6F_6U;

kmatrix(26,26) = -(2*k_5U_4H_1F + 2*k_5U_3H_2F + k_5U_5D);
kmatrix(26,14) = k_5C_5U;
kmatrix(26,20) = k_5F_5U;

kmatrix(27,27) = -(2*k_4U_3H_1F + k_4U_2H_2F + k_4U_4D);
kmatrix(27,15) = k_4C_4U;
kmatrix(27,21) = k_4F_4U;

kmatrix(28,28) = -(2*k_3U_2H_1F + k_3U_3D);
kmatrix(28,16) = k_3C_3U;
kmatrix(28,22) = k_3F_3U;

kmatrix(29,29) = -(k_2U_1H_1F + k_2U_2D);
kmatrix(29,17) = k_2C_2U;
kmatrix(29,23) = k_2F_2U;

kmatrix(30,30) = -(k_1U_1D);
kmatrix(30,18) = k_1C_1U;
kmatrix(30,24) = k_1F_1U;

kmatrix(31,31) = -(k_6D_5F_1F + k_6D_4F_2F + k_6D_3F_3F);
kmatrix(31,25) = k_6U_6D;

kmatrix(32,32) = -(k_5D_4F_1F + k_5D_3F_2F);
kmatrix(32,26) = k_5U_5D;

kmatrix(33,33) = -(k_4D_3F_1F + k_4D_2F_2F);
kmatrix(33,27) = k_4U_4D;

kmatrix(34,34) = -(k_3D_2F_1F);
```

```
kmatrix(34,28) = k_3U_3D;

kmatrix(35,35) = -(k_2D_1F_1F + k_2D_2O);
kmatrix(35,29) = k_2U_2D;

kmatrix(36,36) = -k_1D_1O;
kmatrix(36,30) = k_1U_1D;

kmatrix(41,41) = -k_2O_1O_1O;
kmatrix(41,35) = k_2D_2O;

kmatrix(42,42) = -k_1O_1S;
kmatrix(42,36) = k_1D_1O;
kmatrix(42,41) = 2*k_2O_1O_1O;


kmatrix(48,42) = k_1O_1S;
```

Supplementary Material S5

Table of the kinetic constants obtained by fitting procedure is provided as an Excel file.