

Supplementary document file

**Reversible transformation of a vesicular aggregate in response to pH oscillation**

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### Analytical solution of Eq. 1

The solutions to Eq. 1 for conditions (i), (ii), (i'), and (ii') are as follows.

$$\frac{dx}{dt} = a\{f(t) - x\} \quad (1)$$

For (i) and (ii), Laplace transform may be applied.

$$X(s) = \frac{x(0)}{s+a} + \frac{a}{s+a} F(s),$$

where  $X(s)$  and  $F(s)$  are the Laplace transforms of  $x(t)$  and  $f(t)$ , respectively.

(i)  $f(t) = 0, t < 0$  and  $t > \tau; = K, 0 < t < \tau$

$$F(s) = \int_0^{\tau} K e^{-st} dt = \frac{K(1 - e^{-s\tau})}{s}$$

As  $x(0) = 0$ , the inverse Laplace transform provides

$$x(t) = Ka[(1 - e^{-at}) - H(t - \tau)\{1 - e^{-a(t-\tau)}\}].$$

Here,  $H(t)$  denotes the Heaviside function.

(ii)  $f(t) = 0, t < 0$  and  $t > \tau; = -k(t - \tau), 0 < t < \tau$

$$F(s) = -k \int_0^{\tau} (t - \tau) e^{-st} dt = -\frac{k(1 - e^{-s\tau})}{s^2} + \frac{k\tau}{s}$$

As  $x(0) = 0$ , the inverse Laplace transform provides

$$x(t) = \frac{k(1+a\tau)}{a} (1 - e^{-at}) - \frac{k}{a} H(t - \tau) (1 - e^{-a(t-\tau)}) - k\{t - R(t - \tau)\}.$$

Here,  $R(t)$  denotes the Ramp function

$$R(t) = t \text{ at } t > 0; = 0 \text{ at } t < 0.$$

For (i') and (ii'), Fourier series may be applied.

(i')  $f(t) = 0$  at  $-\tau < t < -\frac{\tau}{2}$  and  $\frac{\tau}{2} < t < \tau,; = K$  at  $-\frac{\tau}{2} < t < \frac{\tau}{2}$

$$x(t) = \sum_{n=-\infty}^{\infty} c_n e^{i\frac{n\pi t}{\tau}}, \quad f(t) = \sum_{n=-\infty}^{\infty} f_n e^{i\frac{n\pi t}{\tau}},$$

where  $c_n$  and  $f_n$  are complex Fourier coefficients of  $x(t)$  and  $f(t)$ , respectively.

Applying the Fourier series to Eq. 1 yields

$$c_n = \frac{af_n}{a + i\frac{n\pi}{\tau}}.$$

Here,

$$c_n = \frac{1}{2\tau} \int_{-\tau}^{\tau} x(t) e^{-i \frac{n\pi t}{\tau}} dt, \quad f_n = \frac{1}{2\tau} \int_{-\tau}^{\tau} f(t) e^{-i \frac{n\pi t}{\tau}} dt$$

These yield

$$x(t) = \sum_{n=-\infty}^{\infty} \frac{a}{a + i \frac{n\pi}{\tau}} \frac{K}{n\pi} \sin \frac{n\pi}{2} e^{i \frac{n\pi t}{\tau}}.$$

Using trigonometric function,  $x(t)$  becomes

$$x(t) = \frac{K}{2} + \sum_{n=1}^{\infty} \frac{K}{n\pi} \sin \frac{n\pi}{2} \frac{2a}{a^2 + \left(\frac{n\pi}{\tau}\right)^2} \left( a \cos \frac{n\pi t}{\tau} + \frac{n\pi}{\tau} \sin \frac{n\pi t}{\tau} \right).$$

(ii')  $f(t) = 0$  at  $-\tau < t < 0$ ;  $= -k(t - \tau)$   $0 < t < \tau$

The same procedure for the given  $f(t)$  yields

$$x(t) = \frac{k\tau}{4} + \frac{1}{2\tau} \sum_{n=1}^{\infty} \frac{a}{a^2 + \left(\frac{n\pi}{\tau}\right)^2} \left[ \frac{2k\tau^2}{n\pi} \left\{ a \sin \frac{n\pi t}{\tau} - \frac{n\pi}{\tau} \cos \frac{n\pi t}{\tau} \right\} - k \left( \frac{\tau}{n\pi} \right)^2 (\cos n\pi - 1) \left\{ 2a \cos \frac{n\pi t}{\tau} + \frac{2n\pi}{\tau} \sin \frac{n\pi t}{\tau} \right\} \right].$$