Lecture Note 6

Depletion Methods

Nov. 2008

Prof. Joo Han-gyu
Department of Nuclear Engineering

Fission Product Poisoning

- Xenon properties

$$\sigma_{th} = 2.72\times10^{6} b \quad (cf.\ \sigma_{th}^{U-235} = 230b)$$

$$\begin{tikzcd}
Te-135 \arrow[r, "\beta{,}11sec"'] & I-135 \arrow[r, "\beta{,}6.7hr"'] & Xe-135 \arrow[r, "\beta{,}9.2hr"'] & Cs-135 \arrow[r, "\beta{,}2.3\times10^{6}hr"'] & Ba-135 \\
Fission \arrow[u] & Fission\ (0.0639) \arrow[u] & Fission\ (0.00237) \arrow[u] & &
\end{tikzcd}$$

- Samarium properties

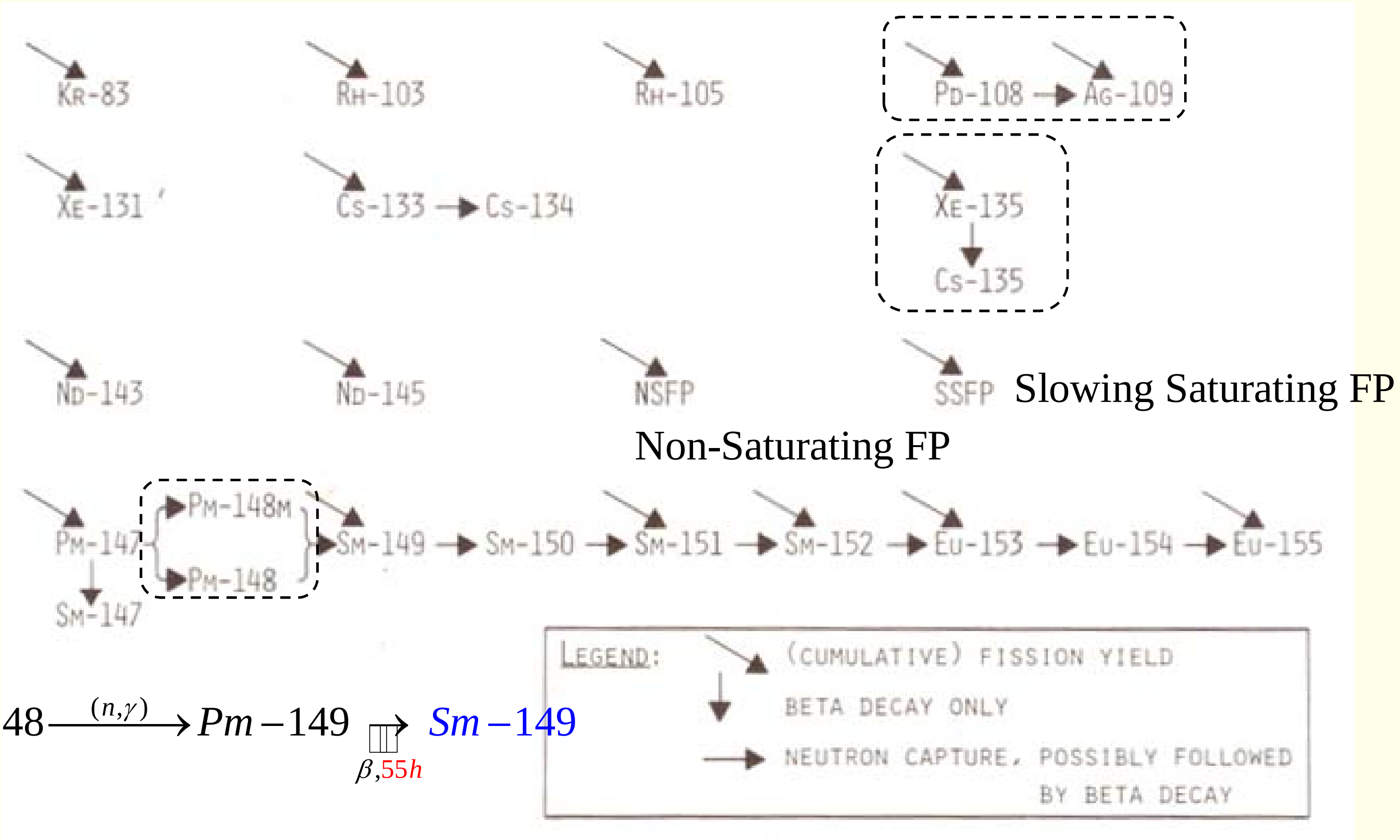
$$\sigma_{th} = 40800b$$

$$\begin{tikzcd}
Nd-149 \arrow[r, "\beta{,}1.8h"'] & Pm-149 \arrow[r, "\beta{,}55h"'] & Sm-149 \\
Fission \arrow[u] & Fission\ (0.01071) \arrow[u] &
\end{tikzcd}$$

Because of short half lives and small fission yield, $Te-135$ and $Nd-149$ are merged with I-135 and Pm-149, respectively.
*$Barium, Neodymium, Promethium$

Proper estimation of the FP content is very important for assessing criticality.

Fission Product Chain

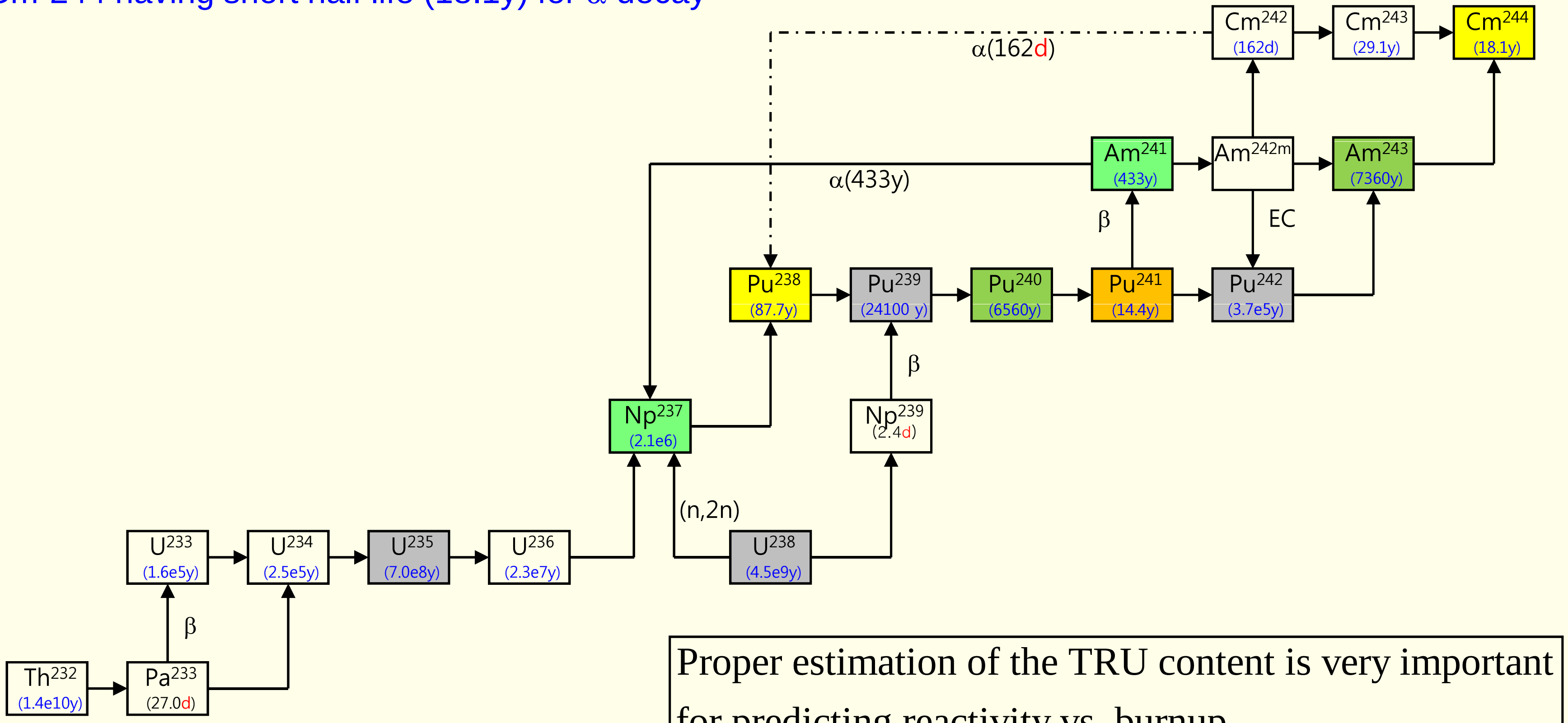


$$Pm-148 \xrightarrow{(n,\gamma)} Pm-149 \xrightarrow[\beta,55h]{} Sm-149$$

Various modes of production (fission, capture, decay) and extinction (absortion, decay)

Fuel Burnup Chain

- Pu-239 from capture in U-238, successive capture for other Pus
- Am-241 from beta decay of Pu-241 (14.4 y)
- Np-237 initially from U-235 during burnup, later from α decay of Am-241
- Cm-244 having short half life (18.1y) for α decay



Proper estimation of the TRU content is very important for predicting reactivity vs. burnup because they are fissionable or even fissile.

Depletion Equation and Its Solution

$$\frac{dX_i(t)}{dt} = \sum_{\substack{j=1\\ j\neq i}}^{N} \ell_{ij}\lambda_j X_j + \bar{\phi}\sum_{\substack{j=1\\ j\neq i}}^{N} \gamma_{ij}\sigma_j X_j - (\underbrace{\lambda_i + \sigma_i\bar{\phi}}_{d_i})X_i \quad (i = 1,\ldots,N)$$: Bateman Equation

$X_i(t)$: atomic density of nuclide i
λ_i : decay constant of nuclide i
ℓ_{ij} : yield fraction of nuclide i from decay of nuclide j
γ_{ij} : yield fraction of nuclide i from reaction of nuclide j
$\bar{\phi}$: position- and energy-averaged flux (1group)
σ_i : spectrum-averaged absorption cross section of nuclide i (1group)
d_i : effective decay constant $(\lambda_i + \sigma_i\bar{\phi})$

- Matrix Form

$$\frac{d\mathbf{x}(t)}{dt} = \mathbf{A}\mathbf{x}(t), \quad \mathbf{x}(0)\,\text{given}$$

$$\mathbf{A} = [a_{ij}]$$

$$a_{ii} = -d_i$$
$$a_{ij} = \ell_{ij}\lambda_j + \gamma_{ij}\sigma_j\bar{\phi}, \quad \forall j \neq i$$

–Trial solution: $\mathbf{x}(t) = \mathbf{u}e^{\lambda t}, \quad \mathbf{u} = [u_1, u_2 \cdots, u_n]^T$

$$\frac{d\mathbf{x}(t)}{dt} = \mathbf{u}\lambda e^{\lambda t} = \mathbf{A}\mathbf{u}e^{\lambda t} \rightarrow \mathbf{A}\mathbf{u} = \lambda\mathbf{u}$$

n eigenvalues and eigenvectors of $\mathbf{A}: \lambda_i, \mathbf{u}_i \quad \forall i = 1 \cdots n$

–General solution: $\mathbf{x}(t) = \sum_{i=1}^{n} c_i \mathbf{u}_i e^{\lambda_i t}$

$$-IC: \mathbf{x}(0) = \sum_{i=1}^{n} c_i\mathbf{u}_i \quad = \mathbf{S}\mathbf{c} \;\leftarrow \mathbf{S} = [\mathbf{u}_1, \mathbf{u}_2, \cdots \mathbf{u}_n],\ \mathbf{c} = [c_1, \cdots c_n]^T$$

$$\rightarrow \mathbf{c} = \mathbf{S}^{-1}\mathbf{x}(0)$$

Matrix Exponential

- Matrix form of general solution:

$$\mathbf{x}(t)=\sum_{i=1}^{n} c_i\mathbf{u}_i e^{\lambda_i t}=[\mathbf{u}_1,\mathbf{u}_2\cdots,\mathbf{u}_n]\begin{bmatrix}c_1e^{\lambda_1 t}\\ c_2e^{\lambda_2 t}\\ \vdots\\ c_ne^{\lambda_n t}\end{bmatrix}=\mathbf{S}\underbrace{\begin{bmatrix}e^{\lambda_1 t}&&&\\ &e^{\lambda_2 t}&&\\ &&\ddots&\\ &&&e^{\lambda_n t}\end{bmatrix}}_{\mathbf{D}(t)}\begin{bmatrix}c_1\\ c_2\\ \vdots\\ c_n\end{bmatrix}=\mathbf{SD}(t)\mathbf{S}^{-1}\mathbf{x}(0)$$

- Define matrix exponential:

$$e^{\mathbf{A}t}=\mathbf{S}\begin{bmatrix}e^{\lambda_1 t}&&&\\ &e^{\lambda_2 t}&&\\ &&\ddots&\\ &&&e^{\lambda_n t}\end{bmatrix}\mathbf{S}^{-1} \qquad \to \text{in general}: f(A)=\mathbf{S}diag(f(\lambda_i))\mathbf{S}^{-1}$$

- Alternate derivation using Taylor expansion

$$y'=ay\to y=e^{at}$$

$$y=1+at+\frac{a^2t^2}{2}+\frac{a^3t^3}{3!}+\cdots\frac{a^nt^n}{n!}+$$

$$y'=a+a^2t+\frac{a^3}{2!}t^2+\cdots\frac{a^nt^{n-1}}{(n-1)!}+$$

$$=a(1+at+\frac{a^2t^2}{2}+\frac{a^3t^3}{3!}+\cdots\frac{a^nt^n}{n!}+)=ay$$

For $\mathbf{x}'=\mathbf{A}\mathbf{x}$, let $\mathbf{x}(t)=\sum_{k=0}^{\infty}\frac{(\mathbf{A}t)^k}{k!} \quad \equiv e^{\mathbf{A}t}$

$$\mathbf{x}'=\sum_{k=1}^{\infty}\frac{\mathbf{A}^k t^{k-1}}{(k-1)!}=\mathbf{A}\sum_{k=1}^{\infty}\frac{(\mathbf{A}t)^{k-1}}{(k-1)!}=\mathbf{A}\sum_{k=0}^{\infty}\frac{(\mathbf{A}t)^k}{k!}=\mathbf{A}\mathbf{x}(t)$$

$$\to\mathbf{x}(t)=e^{\mathbf{A}t}\mathbf{x}(0)$$

$$e^{\mathbf{A}t}=\mathbf{I}+\mathbf{A}t+\frac{\mathbf{A}^2}{2}t^2+\frac{\mathbf{A}^3}{3!}t^3+\cdots+\frac{\mathbf{A}^n}{n!}t^n+\cdots$$

Evaluation of Matrix Exponential

$$\mathbf{x} = e^{\mathbf{A}t}\mathbf{x}(0) = \left(\mathbf{I} + \mathbf{A}t + \frac{\mathbf{A}^2}{2}t^2 + \frac{\mathbf{A}^3}{3!}t^3 + \cdots + \frac{\mathbf{A}^n}{n!}t^n\right)\mathbf{x} = \sum_{k=0}^{n}\mathbf{x}_k$$

where $\mathbf{x}_k = \dfrac{A^k}{k!}\mathbf{x}(0)$ $\quad \rightarrow \mathbf{x}_k = \dfrac{1}{k}\mathbf{A}\mathbf{x}_{k-1}$ $\quad \rightarrow$ No need for constructing $\mathbf{A}^k$!

- when to terminate? or which n?

- Taylor Series of Exponential Function

$$e^x = 1 + x + \frac{1}{2}x^2 + \frac{1}{3!}x^3 + \cdots + \frac{1}{n!}x^n + \cdots$$

Would the terms be monotonously decreasing?

- Maximum term in the Taylor series

Let m be the interpart of x, then

$\dfrac{x^m}{m!}$ is the largest term. (decreasing beyond this term due to large value of n!)

$$pf)\qquad \frac{x^{m+1}}{(n+1)!} = \frac{x^m}{n!}\cdot\frac{x}{n+1} < \frac{x^m}{n!}\cdot\frac{n+1}{n+1} = \frac{x^m}{n!} \qquad \because n \le x < n+1$$

$$\frac{x^{m-1}}{(n-1)!} = \frac{x^m}{n!}\cdot\frac{n}{x} < \frac{x^m}{n!}\cdot\frac{n}{n} = \frac{x^m}{n!}$$

Evaluation of Exponential Function with Finite Arithemetic

- Consider an exponential function with a negative argument

$$e^{-x} = 1 - x + \frac{1}{2}x^2 - \frac{1}{3!}x^3 + \cdots + \frac{(-1)^n}{n!}x^n + \cdots$$

- Double precision arithemetic (16 significant digits) for calculation of e^{-x} leading to a small number e.g. 10^{-6}

$$e^{-x} = 10^{-6} \rightarrow x = 6\ln 10 = 13.8155$$

  - The largest term to be added, $m = 13$

$$\frac{13^{13}}{13!} = 48638.84164 = \underbrace{48638841640}_{11\text{digits}} \times 10^{-6}$$

  - Remaining digits for value around 10^{-6}: 5 digits
  - For larger $x > 13.8$, fewer significant digits will be available for e^{-x}
  - In order to retain 5 significant digits at least, $|x| < 13.8$

- In origin 2, 10^{-3} is the reference value

$$e^{-x} = 10^{-3} \rightarrow x = 3\ln 10 = 6.9077$$

$$\frac{6^6}{6!} = 64.8 = \underbrace{64800}_{5\text{digits}} \times 10^{-3} \rightarrow 11\text{digits remaining}$$

Evaluation of Exponential Function with Finite Arithemetic

- what if $|x| > 13.8$?

  1) Keep dividing x by 2 until $|\tau| < 13.8$.

  $\rightarrow k$ time division: $\tau = \frac{x}{2^k} \quad \rightarrow \quad e^{-x} = e^{-\tau \cdot 2^k} = (((e^{-\tau})^2)^2)^2 \ldots)^2$

  2) Obtain $\alpha = e^{-\tau}$ by Taylor series

  3) Take the square of α k times

- Number of terms needed for convergence

  – Error of the k – th term in the Taylor series if neglected $\varepsilon_k(x) = \frac{\frac{x^k}{k!}}{e^{-x}}$

  – number of terms needed for achive $\varepsilon_k(x) < 0.001$

  $$k_{0.1\%} = k(x) = \frac{7}{2}x + 5 \text{ around } x = 13$$

  ~ 40 terms are needed

x	k
10	40
11	43
12	47
13	50
14	54
15	57

- Matrix Norm for association with scalar in Matrix Exponential

$$\|\mathbf{A}\delta t\| = min\{max_i \sum_j |a_{ij}\delta t|, max_j \sum_i |a_{ij}\delta t|\}$$

Simplification of Decay Chains

- Problems associated with short lived nuclides

$$\rightarrow \text{ small } \tau_{1/2} \rightarrow \text{large } \lambda \rightarrow \text{Large norm of } \mathbf{A} \rightarrow \begin{cases} \text{Multiple Halving/Squaring} \\ \text{More terms in expansion} \end{cases}$$

- Removal of short lived nuclides in the chain

$$A \xrightarrow{\lambda_A} B \xrightarrow{\lambda_B} C \Rightarrow A \xrightarrow{\lambda_A} C \quad \text{if } \lambda_A << \lambda_B$$

$$A \xrightarrow{\lambda_A} B \xrightarrow{\lambda_B} C \Rightarrow B \xrightarrow{\lambda_B} C \quad \text{if } \lambda_A >> \lambda_B \text{ with } \gamma'_B = \gamma_A + \gamma_B$$

- Evaluation of matrix exponential with long lived ones only

  – Criterion for Long Lived ones

$$(\lambda_i + \sigma_i \bar{\phi})\delta t < \underbrace{6.9077}_{3\ln 10} \rightarrow \frac{\ln 2}{t_{1/2}} \delta t < 6.9077 \quad \rightarrow \frac{\ln 2}{6.9077} \delta t < t_{1/2} \rightarrow 0.1003 \delta t < t_{1/2} \rightarrow \frac{1}{10} \delta t < t_{1/2}$$

$$t_{1/2} \equiv \frac{\ln 2}{\lambda_i + \sigma_i \bar{\phi}}$$

Solution of Bateman Equation
for Forward Branching Chain for Short Lived Isotopes

$$X_1 \to X_2 \to X_3 \cdots \to X_n$$

$$\left.\begin{matrix} X_A \\ X_B \end{matrix}\right\} \to X_2 \to X_3 :$$

Linearization

$$X_A \to X_2 \to X_3$$

$$X_B \to X_2 \to X_3$$

$$\frac{dX_i(t)}{dt} = a_{i,i-1}X_{i-1} - d_i X_i \text{ : only one precursor}$$

$$i=1: \frac{dX_1(t)}{dt} = -d_1 X_1 \to X_1(t) = X_1(0)e^{-d_1 t}$$

$$i=2: \frac{dX_2(t)}{dt} = -d_2 X_2 + a_{21}X_1$$

$$\frac{d}{dt}\left(X_2(t)e^{d_2 t}\right) = a_{21}X_1(0)e^{(d_2-d_1)t}$$

$$X_2(t)e^{d_2 t} - X_2(0) = a_{21}X_1(0)\frac{e^{(d_2-d_1)t}-1}{d_2-d_1}$$

$$X_2(t) = X_2(0)e^{-d_2 t} + a_{21}X_1(0)\frac{e^{-d_1 t}-e^{-d_2 t}}{d_2-d_1}$$

$$i=3: \frac{dX_3(t)}{dt} = -d_3 X_3 + a_{32}X_2$$

$$\frac{d}{dt}\left(X_3(t)e^{d_3 t}\right) =$$

$$a_{32}\left(X_2(0)e^{(d_3-d_2)t} + a_{21}X_1(0)\frac{e^{(d_3-d_1)t}-e^{(d_3-d_2)t}}{d_2-d_1}\right)$$

$$X_3(t) = X_3(0)e^{-d_3 t} + a_{32}X_2(0)\frac{e^{-d_2 t}-e^{-d_3 t}}{d_3-d_2}$$

$$+\boxed{\frac{a_{32}a_{21}X_1(0)}{d_2-d_1}\left(\frac{e^{-d_1 t}-e^{-d_3 t}}{d_3-d_1} - \frac{e^{-d_1 t}-e^{-d_3 t}}{d_3-d_2}\right)}$$

$$\downarrow$$

$$\to X_1(0)a_{32}a_{21}\left(\frac{e^{-d_1 t}-e^{-d_3 t}}{d_3-d_1}\frac{1}{d_2-d_1} + \frac{e^{-d_1 t}-e^{-d_3 t}}{d_3-d_2}\frac{1}{d_1-d_2}\right)$$

$$X_i(t) = X_i(0)e^{-d_i t}$$

$$+\sum_{k=1}^{i-1} X_k(0)\prod_{n=k}^{i-1} a_{n+1,n}\left[\sum_{j=k}^{i-1}\frac{e^{-d_j t}-e^{-d_i t}}{d_i-d_j}\prod_{\substack{n=k \\ n\neq j}}^{i-1}\frac{1}{d_n-d_j}\right]$$

Solution of Bateman Equation
for Forward Branching Chain for Short Lived Isotopes

$$X_i(t) = X_i(0)e^{-d_i t}$$

$$+\sum_{k=1}^{i-1} X_k(0)\prod_{n=k}^{i-1}\frac{a_{n+1,n}}{d_n}\left[\sum_{j=k}^{i-1} d_j \frac{e^{-d_j t} - e^{-d_i t}}{d_i - d_j}\prod_{\substack{n=k \\ n\neq j}}^{i-1}\frac{d_n}{d_n - d_j}\right]$$