Exact solution for two-phase non-equilibrium mass transfer of air sparging

Osama Al-Gahtani\textsuperscript{1}, James Warner, Paul DuChateau
Colorado State University

Abstract. The exact solution is presented here to evaluate the non-equilibrium mass transfer at the interface between the dissolved and air phase during air sparging. The solution has been developed for solving the contaminant concentrations in both aqueous phase and air phase. The solution is derived by solving the couple system of differential equations in the dissolved and gas phase. The mass transfer in air sparging is relatively fast and a common assumption is to use Henry’s law to describe the mass transfer at the interface based on assumption of equilibrium condition. In this solution, the mass transfer is described based on non-equilibrium formula of two-film theory where the mass transfer coefficient and surface area are included. Also, groundwater advection is considered where most air sparging model assumed that the dissolved phase is stationary due to slow movement of groundwater. In addition, the solution is valid for air channel and for discrete air bubbles where the surface area is different in both forms. Finally, the exact solution was compared with numerical solution and they matched well at all points on tested domain.

1. Introduction.

Water pollution due to industrials activities is always associated with multiphase flow. For remediation purpose and for groundwater contaminated with volatile organic compound, the contaminant may be extracted by stripping the contaminated water. This process is either done by in-situ treatment (air sparging, in-well air stripping) or above ground treatment (air stripping tower, shallow trays etc). In all these technologies, air has to introduce to separate the contaminant to the air phase. For such technologies, the rate of the mass transfer between the two phases is a key factor for efficient remediation system.

In modeling of above ground treatment technology, non-equilibrium mass transfer based on two-film theory (Whitman, 1923) is used for design formulas (Kavanaugh and Trussell, 1980). Recently, this theory starts to get wide acceptance (Gvirtzman and Gorelick, 1992, Braida and Ong, 2000, Chao et al., 1998, and Elder et al., 1999) to be used for modeling the in-situ remedial technology of air sparging. However, there are different conditions where air introduces to enhance the volatilization process. Air can be flowed in an opposite direction of contaminated groundwater (counter-current) and this is what the case for operating air stripping tower. Air can be set in crossing the groundwater
flow and this is the case of cross-flow in-suit air sparging (Pankow et al, 1993, Al-Gahtani, and Warner, 2006). Al-Gahtani, and Warner, 2006 derived the linked differential equations for the cross flow air sparging and developed the numerical solution using finite difference technique for solving the contaminant concentrations at the dissolve and air phases. The numerical solution was compared with analytical formula developed for this study.

In air sparging, there are three main mechanisms to remove the contaminant in the source and dissolved zones using air sparging (Figure 1). The first mechanism is volatilization to remove the hydrocarbons present in the dissolved phase by vaporization of the contaminant from the dissolved to the air phase. The second mechanism is direct vaporization from the hydrocarbon phase to the air phase, which exists in the source zone. The final process is enhanced biodegradation in the dissolved phase, which is achieved by increasing the oxygen content. The contaminant dissolved from source to the dissolved zone can be removed either by volatilization or biodegradation. Experimental and field studies done by Johnson, (1998), Johnston et al., (1998), and Aelion and Kirtland, (2000), showed that biodegradation is much less effective than volatilization. Mass removal from the dissolved zone by volatilization is the focus of this study. McCray (2000) investigates the different form of each phase for air sparging. This study investigates the mass transfer between the dissolved and air phase by volatilization process.

Figure 1: Mechanisms control the air sparging remediation process.

The analytical solution for multiphase flow is always attractive to designers as it gives an easy and simple solution. In most cases, the required assumptions for the analytical solutions are homogenous with simple boundary conditions. For multiphase flow, finding the exact solution is always challenging task because there are more than one equation needs to be solved for the same number of unknowns. Most analytical solutions for air sparging are developed based on the assumption of one phase, a completely mixed zone, and equilibrium partitioning from one phase to another (Sellers, K.L., and R. P. Schreiber, 1992, Wilson et al, 1992, Pankow et al, 1993). The analytical solution presented in this chapter is not based on the above assumptions, and it also includes the fluxes of the water and air phases as they cross each other.
2. Governing Equations

A simulation of two-dimensional flow is considered for an air sparging system (Figure 2) where the air flow direction crosses the groundwater current. The boundary condition of the system is simulated for an air sparging trench problem to approximate an air sparging wells series. At the lower boundary of the system, clean air with zero concentration of contaminant is injected with a constant rate. Contaminated groundwater introduced from the left side of the trench has a constant concentration along the height. The governing differential equations for two phase system in air and dissolved are developed by looking at the contaminant derived to the micro scale elements of representative element volume (REV).

![Figure 2: Boundary conditions of air sparging system for trench type](image)

Using mass balance of the concentrations change in the dissolved and gas phases, the differential equations and boundary conditions of the concentration in the dissolved and gas phase (Figure 2) can be written as (Al-Gahtani, 2006):

\[
\frac{\partial C}{\partial x} = -K_1 (C - C^*) \quad C(0,y) = C_o \quad \text{(1)} \quad \text{and} \quad \frac{\partial C^*}{\partial y} = K_2 (C - C^*) \quad C^*(x,0) = 0 \quad \text{(2)}
\]

Where \( K_1 = K_L a / q_w \) and \( K_2 = K_L a / (q_{air} H) \),

- \( q_{air} \) = air flux and it can be approximated as \( q_{air} = Q_{air} / (\text{Length} \times \text{Width}) \)
- \( Q_{air} \) = Airflow rate in \( \text{L}^3/\text{T} \).
- \( q_w \) = the groundwater velocity it can be approximated as \( q_w = Q_w / (\text{height} \times \text{width}) \)
- \( Q_w \) = flow rate of water effluent into system.
- \( K_L \) = Overall mass transfer coefficient.
- \( a \) = surface area of gas boundary per bulk volume.
- \( C \) = Concentration of contaminant in dissolved phase.
- \( C^* \) = Equivalent dissolve concentration in air phase (Mass/Volume).

In most air stripping applications in chemical and environmental engineering, \( K_L a \) is combined in one parameter factor and evaluated from the experimental fitting. From available data, \( K_L a \) is considered as two separated variables.
3. Analytical Derivations

The equations (1) and (2) are a coupled system of first order linear partial differential equations with two starting boundary conditions. This system behaves essentially like two ordinary differential equations that are weakly coupled through the solution. Since each equation applies on a semi infinite interval and is subject to an initial boundary condition, the Laplace transform can be used to solve these equations. Transforming using both variables reduces the system to a system of two linear algebraic equations, but there is then some difficulty in inverting.

On the other hand, transforming in just one variable or the other leads to a simple ordinary differential equation problem in the untransformed variable. In order to easily invert the transform, it is necessary to use a Taylor series to expand the exponential term which appears in the solution of the ordinary differential equation. Whether transforming in $x$ or $y$ seems to make little difference, and both approaches are shown here. Note also that changing variables eliminates the parameters $K_1$ and $K_2$ from the two equations. The solution will be as functions of $K_1x$ and $K_2y$, and the parameters $K_1$ and $K_2$ describe the soil and fluids properties.

3.1 Solution Using Laplace Transformation in $x$ Direction

Letting $C_f(s, y) = L_s[C(x, y)]$ and $C^*_f(s, y) = L_s[C^*(x, y)]$ are Laplace transformed of the independent variables $C$ and $C^*$, the Laplace transform of equation (1) in $x$ direction and solving $C_f$ in terms $C^*_f$ as

$$C_f(s, y) = \frac{C_o}{s + K_1} + \frac{K_1}{s + K_1}C_i^*(s, y) \quad (3)$$

Now the Laplace transform of equation (1) will be

$$\frac{\partial C_f^*(s, y)}{\partial y} = K_2(C_f - C_f^*) \quad (4)$$

The transform of the boundary condition of equation (2) is

$$C_f^*(s, 0) = L_s[C^*(x, 0)] = 0 \quad (5)$$

The algebraic combination of equation (3) and (4) becomes

$$\frac{\partial C_f^*(s, y)}{\partial y} = -\frac{K_2s}{s + K_1}C_f^*(s, y) + \frac{K_2C_o}{s + K_1} \quad (6)$$

Solving the ordinary differential equation of (6) with boundary condition of equation (5) will get
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\[ C_i'(s, y) = \frac{C_o}{s} \left[ 1 - e^{-K_{22} \frac{s}{s + K_i}} \right] \]  \hspace{1cm} (7)

Using the expansion of exponential series (i.e. \( e^y = \sum_{n=0}^{\infty} \frac{y^n}{n!} \)) of equation (7) and substituting that into equation (3), the term \( C_i'(s, y) \) becomes

\[ C_i(s, y) = \frac{C_o}{s + K_i} - K_i C_o \sum_{n=1}^{\infty} \frac{(-K_{22} y)^n}{n!} \frac{s^{n-1}}{(s + K_i)^{n+1}} \]  \hspace{1cm} (8)

Since

\[ L_x^{-1} \left[ \frac{1}{(s + a)^y} \right] = \frac{1}{(s - 1)^y} x^{n-1} e^{-K_{12} x} \quad \text{and} \quad L_x^{-1} \left[ \frac{s^{n-1}}{(s + K_i)^{n+1}} \right] = \left( \frac{d}{dx} \right)^{n-1} \left[ \frac{1}{n!} x^n e^{-K_{12} x} \right] \]

The inverse of Laplace will hold the concentration of the contaminant in the aqueous phase in the form of

\[ C(x, y) = C_o e^{-K_{12} x} + C_o \sum_{n=1}^{\infty} \frac{(-K_{22} y)^n}{n!} \phi_n(K_i x) (-K_i x) e^{-K_{12} x} \]

where

\[ L_x^{-1} \left[ \frac{1}{(s + K_i)^y} \right] = x e^{-K_{12} x} = \phi_0(K_i x) x e^{-K_{12} x} \]

\[ L_x^{-1} \left[ \frac{s}{(s + K_i)^y} \right] = \frac{1}{2} \frac{d}{dx} x^2 e^{-K_{12} x} = \left( 1 - \frac{1}{2} K_i x \right) x e^{-K_{12} x} = \phi_1(K_i x) x e^{-K_{12} x} \]

\[ L_x^{-1} \left[ \frac{s^2}{(s + K_i)^y} \right] = \frac{1}{3} \frac{d^2}{dx^2} x^3 e^{-K_{12} x} = \left( 1 - K_i x + \frac{1}{6} (K_i x)^2 \right) x e^{-K_{12} x} = \phi_2(K_i x) x e^{-K_{12} x} \]

and in general

\[ L_x^{-1} \left[ \frac{s^{n-1}}{(s + K_i)^y} \right] = \frac{1}{(n-1)!} \frac{d^{n-1}}{dx^{n-1}} x^n e^{-K_{12} x} = \phi_{n-1}(K_i x) x e^{-K_{12} x} \quad \text{for } n = 1, 2, \ldots \]  \hspace{1cm} (9)

\( \phi_m(x) \) is a polynomial of degree \( m \) in \( x \) such that \( \phi_m(0) = 1 \) for every \( m \) then
\[ C(x, y) = C_0 e^{-K_1 x} \left[ 1 + \sum_{n=1}^{\infty} \frac{(-K_2 y)^n}{n!} (-K_1 x) \phi_{n-1}(K_1 x) \right] \] (10 a)

or
\[ C(x, y) = C_0 e^{-K_1 x} \left[ \Phi \left( K_1 x, K_2 y \right) \right] \] (10 b)

where
\[ (K_1 x, K_2 y) = \sum_{n=1}^{\infty} \frac{(-K_2 y)^n}{n!} (-K_1 x) \phi_{n-1}(K_1 x) \] (11)

The \( \Phi \) function evaluation is shown in appendix A. \( \Phi \) function as presented in equation (11) has stability value up to the \( K_2 y \) value of 20. Another way evaluating \( \Phi \) as shown in the following equation:

\[ (K_1 x, K_2 y) = \sum_{n=1}^{\infty} \frac{(-K_2 y)^n}{n!} \sum_{m=0}^{n-1} \frac{(n-1)(-K_1 x)^{m+1}}{(m+1)} \] (12)

3.2 Solution Using the Laplace Transform in \( y \) Direction

Let \( C_i(s, y) = L_y[C(x, y)] \) and \( C_i^+(s, y) = L_y[C^+(x, y)] \)
The Laplace formula in \( y \) direction of equation (2) will be as
\[ C_i^+(x, s) = \frac{K_2}{s + K_2} C_i(x, s) \] (13)

and equation (1) with the boundary condition will be
\[ \frac{\partial C_i(x, s)}{\partial x} = -K_1 \left( C_i(x, s) - C_i^+(x, s) \right) \] (14)

and from the boundary condition
\[ C_i(0, s) = \frac{C_o}{s} \] (15)

Combing equation (13) and (14) becomes
\[ \frac{\partial C_i(x, s)}{\partial x} = -\frac{K_1 s}{s + K_2} C_i(x, s) \] (16)

Solving the ordinary differential equation of equation (16) with boundary condition of equation (15) will lead to
\[ C_i(x, s) = \frac{C_0}{s} e^{-K_{s_1} \left( \frac{s}{s+K_2} \right)} \] (17)

The expansion of the exponential term will be

\[ e^{-K_{s_1} \left( \frac{s}{s+K_2} \right)} = \sum_{n=0}^{\infty} \frac{(-K_{s_1}x)^n}{n!} \left( \frac{s}{s+K_2} \right)^n \] (18)

The transformed concentration in the dissolved phase will be

\[ C_i(x, s) = \frac{C_0}{s} e^{-K_{s_1} \left( \frac{s}{s+K_2} \right)} = C_0 + C_0 \sum_{n=1}^{\infty} \frac{(-K_{s_1}x)^n}{n!} \left( \frac{s}{s+K_2} \right)^n \] (19)

or

\[ C_i(x, s) = \frac{C_0}{s} + C_0 \left[ (-K_{s_1}x) \frac{1}{s+b} + \frac{(-K_{s_1}x)^2}{2!} \left( \frac{s}{s+K_2} \right)^2 + \frac{(-K_{s_1}x)^3}{3!} \left( \frac{s}{s+K_2} \right)^3 + ... \right] \] (20)

Since

\[ L_y \left[ \frac{s^{n-1}}{(s+K_2)^n} \right] = \left( \frac{d}{dy} \right)^{n-1} \left[ \frac{1}{(n-1)!} y^{n-1} e^{-K_{s_1} y} \right] = e^{-K_{s_1} y} \psi_{n-1}(K_{s_1} y) \] (21)

The inverse Laplace will bring the concentration in the dissolved phase in the form of:

\[ C(x, y) = C_0 \left[ 1 + e^{-K_{s_1} y} \sum_{n=1}^{\infty} \frac{(K_{s_1} y)^n}{n!} \psi_{n-1}(K_{s_1} y) \right] \] (22 a)

or

\[ C(x, y) = C_0 \left[ 1 + e^{-K_{s_2} y} \psi(K_{s_1} x, K_{s_2} y) \right] \] (22 b)

where

\[ \psi(K_{s_1} x, K_{s_2} y) = \sum_{n=1}^{\infty} \frac{(K_{s_1} x)^n}{n!} \psi_{n-1}(K_{s_2} y) \] (23)

The equation of (22) is the concentration of the contaminant in the dissolved phase using the Laplace transform in the \( y \) direction. The evolution of the \( \psi \) function as it is presented in equation (23) is shown in Appendix A. In the next section, stability of the equation (12) and (23) will be discussed and general formula to evaluate the polynomial function will be presented.
4 Stability Analysis for the Solutions

This section will focus on studying the stability of equations (12) and (23). Equation (12) is derived from taking the Laplace transform in the \( x \) direction. The \( \Phi \) function evaluated in appendix A. This function has evaluated and given a stable value up to the value of \( K_{1x} \) of 20. Equation (23) is a similar form of evaluating the concentration in the dissolved phase, and it is derived by taking the Laplace transform in the \( y \) direction. By comparing equation (12b) and (23b), the \( \Phi \) function can be written in the following equation.

\[
(K_1x, K_2y) = e^{K_{1x}} \left[ 1 - e^{-K_{2y}} \Psi(K_1x, K_2y) \right] - 1 \quad (24)
\]

where the \( \Psi(K_{1x}, K_{2y}) \) evaluation shown in appendix A.

Another form of equation (24) can be written in binomial terms:

\[
(K_1x, K_2y) = e^{K_{1x}} \left[ 1 - e^{-K_{2y}} \sum_{n=1}^{\infty} \frac{(-K_1x)^n}{n!} \sum_{m=0}^{n} \frac{(-K_2y)^m}{m!} \right] - 1 \quad (25)
\]

The \( \Phi \) function, as they are written in equations (24) and (25), have stability up to the \( K_{2y} \) value of 20. A similar stability form is shown in equation (11) and (12), where \( \Phi \) runs for a stable solution up to \( K_{1x} \) equal to 20. Figure 3 shows the schematic of the stability domain with ranges in both \( x \) and \( y \) directions. From Figure 3, there is a range where the analytical solution can not evaluate. This range includes where both \( K_{1x} \) and \( K_{2y} \) are greater than 20.

The numerical solution developed by Al-Gahtani and Warner, (2006) has a stable solution on a more extended range than does the analytical solution. For taking the advantage of the stability of the numerical solution, \( \Phi \) function can be rewritten in numerical form. Using equation (22b), the third form of \( \Phi \) function based on a numerical solution is

\[
(K_1x, K_2y) = \left( \frac{C}{C_o} \right)_{\text{numerical}} e^{K_{1x}} - 1 \quad (26)
\]

The \( C/C_o \) term of equation (26) can be evaluated by running the numerical program at different \( K_{1x} \) and \( K_{2y} \) values. In this way, \( \Phi \) can be numerically evaluated by generate different \( K_{1x} \) and \( K_{2y} \). The numerical run for evaluating the analytical solution will be for homogenous soil and constant boundary condition. A table of evaluation the \( \Phi \) function at different values of \( K_{1x} \) and \( K_{2y} \) can be generated based on numerical solution.
5 Solution Comparison with Numerical Solution

For comparing the analytical solution of the equation (10), and the numerical solution, a hypothetical case for a site contaminated with TCE is used. The problem consists of a 1 m depth and 2 m length air sparging trench. Site conditions and chemical properties used are summarized in Table 1. The trench is packed with crushed gravel, which assumes that bubbles will form in the trench. The bubble size was assumed to be 1 mm in diameter with bubble velocity of 0.1 m/s.

The concentration profiles are shown in Figure 4 for numerical and analytical results. More details of comparison between the numerical and analytical solutions are shown in group of graphs in figure 5 for different section profiles along the trench’s length and depth. The group of graphs in Figure 7 shows that the numerical and analytical solutions developed in this study are matched well at all the points on the domain.

Table 1: Chemical and soil properties

<table>
<thead>
<tr>
<th>$K_L$ (cm/hr)</th>
<th>$H$</th>
<th>$\phi$</th>
<th>$i$ Gradient</th>
<th>$K$ hydraulic conductivity (cm/s)</th>
<th>$Q_w$ (m$^3$/s)</th>
<th>$Q_{air}$ (m$^3$/s)</th>
<th>air to water ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>20.4</td>
<td>0.42</td>
<td>0.5</td>
<td>0.003</td>
<td>0.001</td>
<td>0.26</td>
<td>0.96</td>
<td>3.7</td>
</tr>
</tbody>
</table>

Figure 4: Program results for numerical and analytical solution
Figure 5: Analytical and numerical developed in chapter 4 comparison along different trench elevations

Figure 6: Analytical and numerical comparison along the trench depth
Appendix A: Evaluating $\Phi$ and $\Psi$ functions

1. Evaluating of $\Phi$ function

The general formula of $\phi$ as shown in equation (9) and rewritten here is

$$
\phi_{n+1}(K_1x) = \frac{1}{xe^{-K_1x}} \frac{1}{(n-1)!} \frac{d^{n-1}}{dx^{n-1}} \left( e^{-K_1x} \right) 
$$

for $n = 1, 2, ...$

By evaluating the derivative to the fourth degree ($n$ up to 5) $\phi$ values will be as:

$\phi_0 = 1$

$\phi_1 = 1 - \frac{1}{2!} (K_1x)$

$\phi_2 = 1 - \frac{1}{2} (K_1x) + \frac{1}{6} (K_1x)^2$

$\phi_3 = 1 - \frac{3}{2} (K_1x) + \frac{1}{2} (K_1x)^2 - \frac{1}{24} (K_1x)^3$

$\phi_4 = 1 - \frac{2}{2} (K_1x) + \frac{1}{6} (K_1x)^2 - \frac{1}{120} (K_1x)^3 + \frac{1}{120} (K_1x)^4$

After rearrange

$\phi_0 = 1$

$\phi_1 = 1 - \frac{1}{2!} (K_1x)$

$\phi_2 = 1 - \frac{2}{2!} (K_1x) + \frac{1}{3!} (K_1x)^2$

$\phi_3 = 1 - \frac{3}{2!} (K_1x) + \frac{3}{3!} (K_1x)^2 - \frac{1}{4!} (K_1x)^3$

$\phi_4 = 1 - \frac{4}{2!} (K_1x) + \frac{6}{3!} (K_1x)^2 - \frac{4}{4!} (K_1x)^3 + \frac{1}{5!} (K_1x)^4$

In matrix form

$$
\begin{bmatrix}
\phi_0 \\
\phi_1 \\
\phi_2 \\
\phi_3 \\
\phi_4 \\
\end{bmatrix} =
\begin{bmatrix}
1 & 0 & 0 & 0 & 0 \\
1 & -1 & 0 & 0 & 0 \\
1 & -2 & 1 & 0 & 0 \\
1 & -3 & 3 & -1 & 0 \\
1 & -4 & 6 & -4 & 1 \\
\end{bmatrix}
\begin{bmatrix}
(K_1x)^0 / 1! \\
(K_1x)^1 / 2! \\
(K_1x)^2 / 3! \\
(K_1x)^3 / 4! \\
(K_1x)^4 / 5!
\end{bmatrix}
$$

As noticed the first column of the matrix has the value of 1 and alternates the sign as marching to the next columns. As shown above, adding any two absolute consequences number in row (dashed circles) gives the second number’s position in column and next
row’s position (solid circle). By this way and using spreadsheet, the middle matrix can be generated to the desired terms. After that the $\Phi$ term as mentioned in equation (9) can be evaluated for any values of $K_1$ and $K_2$ and point location $x$ and $y$.

2. Evaluating of $\Psi$ function.

Evaluating of $\Psi(K_1x,K_2y)$ function

\[
\begin{align*}
\psi_0 &= 1 \\
\psi_1 &= 1 - (K_2y) \\
\psi_2 &= 1 - 2(K_2y) + 1/2(K_2y)^2 \\
\psi_3 &= 1 - 3(K_2y) + 3/2(K_2y)^2 - 1/6(K_2y)^3 \\
\psi_4 &= 1 - 4(K_2y) + 3(K_2y)^2 - 4/6(K_2y)^3 + 1/24(K_2y)^4 \\
\psi_5 &= 1 - 5(K_2y) + 5(K_2y)^2 - 5/3(K_2y)^3 + 5/24(K_2y)^4 - 1/120(K_2y)^5
\end{align*}
\]

After rearrange

\[
\begin{align*}
\psi_0 &= 1 \\
\psi_1 &= 1 - 1/1!(K_2y) \\
\psi_2 &= 1 - 2/1!(K_2y) + 1/2!(K_2y)^2 \\
\psi_3 &= 1 - 3/1!(K_2y) + 3/2!(K_2y)^2 - 1/3!(K_2y)^3 \\
\psi_4 &= 1 - 4/1!(K_2y) + 6/2!(K_2y)^2 - 4/3!(K_2y)^3 + 1/4!(K_2y)^4 \\
\psi_5 &= 1 - 5/1!(K_2y) + 10/2!(K_2y)^2 - 10/3!(K_2y)^3 + 5/4!(K_2y)^4 - 1/5!(K_2y)^5
\end{align*}
\]

In matrix form

\[
\begin{bmatrix}
\psi_0 \\
\psi_1 \\
\psi_2 \\
\psi_3 \\
\psi_4 \\
\psi_5
\end{bmatrix} =
\begin{bmatrix}
1 & 0 & 0 & 0 & 0 & 0 \\
1 & -1 & 0 & 0 & 0 & 0 \\
1 & -2 & 1 & 0 & 0 & 0 \\
1 & -3 & 3 & -1 & 0 & 0 \\
1 & -4 & 6 & -4 & 1 & 0 \\
1 & -5 & 10 & -10 & 5 & -1
\end{bmatrix}
\begin{bmatrix}
(K_2y)^0 \\
(K_2y)^1 \\
(K_2y)^2 \\
(K_2y)^3 \\
(K_2y)^4 \\
(K_2y)^5
\end{bmatrix}
\]

Again the process ends up same middle matrix has been developed for $\Phi$ function as shown in the previous section.
References


