Nonlinear Model Predictive Control of a coagulation chemical dosing unit for water treatment plants *

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Abstract: The need for processes to be operated under tighter performance specifications and satisfy constraints have motivated the increasing applications of nonlinear model predictive control (MPC) by the process industry. Nonlinear MPC conveniently meets the higher product quality, productivity and safety demands of complex processes by taking into account the nonlinear MPC to a multi-variable coagulation chemical dosing unit for water treatment plants. A nonlinear model of the dosing unit based on mechanistic modelling and identified by nonlinear autoregressive with external input (NLARX) estimator was developed. The simulation of the MPC based control system showed very good performance for set-point tracking and disturbance rejection. The closed loop performance of the nonlinear MPC (NMPC) compares favourably with the unconstrained and linearised nonlinear MPC (LTIMPC). The results of this study shows the suitability of nonlinear MPC for process control in the water treatment industry.

1. INTRODUCTION

Coagulation in water treatment plants is a complex and nonlinear process requiring addition of optimum quantity of chemical reagents to raw water to meet the desired standards. One of the key issues in coagulation process is that water quality parameters vary unexpectedly and cannot be manipulated easily. These variations act as perturbation or disturbance to the control loop of the system. The control objective is therefore targeted at manipulating the flow of the coagulants and pH adjustment chemicals to track the set-point signals in the presence of these possibly fast acting disturbances. The traditional control system for coagulation control has been found to have a number of limitations such as inaccurate process model to describe the behaviour of the system, slow responses to longer system delay time, variations in water quality parameters and loop interaction effects within the system.

One of the commonly used control strategies is the feedforward control. It involves adjusting the levels of chemical coagulants added to a process stream as a result of sensory information measured from the raw water variable(s). This is achieved by changing the feed rate of the coagulant metering pump according to the measured flow rate of the raw water [American Water Works Association & American Society of Civil Engineers, 2005]. This approach however becomes inappropriate, when the flow rates vary rapidly and there are large changes in other water quality variables. To address these problems in the feedforward control strategy, several models such as multi-linear regression equations, artificial neural networks and fuzzy inference system algorithms have been proposed to predict the accurate amount of coagulants under varied conditions to replace the influent flowmeter response.

For instance, Evans et al. [1998] proposed a feedforward controller based on adaptive neuro-fuzzy networks for Huntington water treatment works in North West England. In Baxter et al. [2002], the integration of neural network models with the supervisory control and data acquisition (SCADA) system through a number of process optimisation interfaces to optimise the chemical costs and doses online in real-time is presented according to variations in influent water quality parameters. In Fletcher et al. [2002], a feedforward control was developed using models based on nonlinear transformation of variables, multilayer perceptron (MLP) and radial basis function (RBF) network to improve coagulation process. The findings of these research works are positive. However, the application of data-based models with feed-forward controllers to control coagulation process depends on the availability of a perfect model and accurate data from the plant operational records.

Another option in the literature for coagulation control is the application of feedback control strategy. This involves the use of sensors such as streaming current detector to measure the surface charge of the water after the coagulation process, compares the process value with the set point and adjusts the coagulant dosage pump accordingly to correct any deviation from the expected results. It is characterised by a system delay or dead time. During the seasons when the raw water quality changes frequently and widely, the control system may not function effectively re-

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sulting in under dosing or over dosing of coagulation chemicals. In a study on feedback control strategy by Adgar et al. [2005], the authors investigated the application of feedback control on coagulation process on a twin pilot plant using streaming current detector and pH sensor to improve the existing manually flow-proportional coagulant dosage control strategy. Analysis of the data collected during experiments on the pilot plant demonstrated that there is a strong interaction between the streaming current detector and pH measurements. Based on this observation, they proposed a new decoupling control scheme that reduces the interaction between the pH and coagulant dosage loops. The new control strategy was found to be less susceptible to disturbance when compared with the separate feedback control loops. In another study performed by Paz & Ocampo [2009], the author compared the performances of PID and linear model predictive control (MPC) controller for a SISO model of a coagulant dosage system. However, the study does not take into consideration the effect of pH on the coagulation control process.

The combination of feedforward and feedback control to correct the effect of measured disturbances and errors in the system is another strategy that has been proposed. In Hua et al. [2009], a feedforward fuzzy logic controller and feedback controller to determine optimum chemical dosage and to control the coagulant dosage system of a water treatment plant was developed. However, in spite of the attractive nature of fuzzy logic control, the proposed controller has some difficulties, such as knowledge acquisition from experienced operators and large set of rules involved in testing the controller.

This paper therefore examines the application of nonlinear MPC to maintain the controlled variables at the specified reference values or set-points by adjusting the control variables of a multi-input multi-output (MIMO) model of a coagulation chemical dosing unit. The nonlinear MPC is proposed due to its ability to handle nonlinear and multivariable process satisfactorily. It has the ability to predict future events and can take appropriate actions to meet control objective of the plant. It can handle large time delay and higher order dynamics of system model. Nonlinear MPC can accommodate constraints place on the actuator's inputs without driving it to saturation. The coagulation chemical dosing unit at the Rietvlei water treatment plant, South Africa is considered for the study. The study shows the efficacy of nonlinear MPC to control nonlinear and complex process in water treatment plants where traditional control strategies are inadequate or exhibit poor performance.

The paper is organised as follows. Section 2 gives the description of the water treatment plant and the control problem. The process modelling of the chemical dosing unit and model predictive control is discussed in Section 3. The results of different simulation scenarios are presented in Section 4. The concluding remarks are highlighted in the last Section.

2. PLANT DESCRIPTION

Rietvlei water treatment plant was constructed between 1932 and 1934 near Irene, City of Tshwane in South Africa. Fig. 1 shows how the water treatment plant takes raw



Fig. 1. Description of process train at the Rietvlei water treatment plant

waters from the dam through the inlet tower followed by a train of unit processes such as coagulation, flocculation, dissolved air flotation and filtration, granular activated carbon filtration, chlorination and finally distribution to consumers [American Water Works Association & American Society of Civil Engineers, 2005, City of Tshwane, S.a. The reagents for chemical dosing unit in the plant are sudfloc 3835, ferric chloride and hydrated lime. The sudfloc 3835 (mixture of polyamine polymer and aluminium chlorohydrate) and ferric chloride are the primary and secondary coagulants respectively. They neutralise the negative surface charge of the raw waters and destabilised the colloids particles to form flocs that are filtered out as sludge. The hydrated lime is added to adjust the pH level of the waters in the mixing tank so that effective coagulation could take place. The chemical solutions are applied to the mixing tank with the aid of dosing pumps.

The challenge in the water coagulation process is the application of optimum amount of these reagents to the raw waters undergoing treatment in order to meet the laid down standards, satisfy varying water quality and demands. Thus, the control strategy objective is to ensure that the controlled variables of the effluent waters track the reference trajectories of the dosing unit and simultaneously rejects any disturbances arising from variations in operational conditions. This is to be achieved by manipulating the flow rates of the chemical reagents applied to the raw waters in the mixing tank taking into considerations the constraints on the manipulated inputs to prevent the dosing pumps from exceeding their limits.

3. METHODS/MATERIALS

3.1 Nonlinear model of the chemical dosing unit

The coagulation chemical dosing unit involves a nonlinear and physicochemical process. The dynamics of the process model is presented as follows.

The mass balance equations for the mixing tank are [Evangelou, 1998, Gardia & Godoy, 2011, Bello et al., 2013]:

$$V \frac{d \left[(C_5 H_{12} O N^+)_n \right]}{dt} = \left[\left(\left(C_5 H_{12} O N^+ \right)_n \right)_{in} \right] q_a -$$

$$\left[\left(C_5H_{12}ON^+\right)_n\right]q_{out} (1)$$

$$V\frac{d\left[Al^{3+}\right]}{dt} = \left[Al_{in}^{3+}\right]q_a - \left[Al^{3+}\right]q_{out}$$
(2)

$$V\frac{d\left[Fe^{3+}\right]}{dt} = \left[Fe^{3+}_{in}\right]q_b - \left[Fe^{3+}\right]q_{out} \tag{3}$$

$$V\frac{d\left\lfloor Ca^{2+}\right\rfloor}{dt} = \left[Ca_{in}^{2+}\right]q_c - \left[Ca^{2+}\right]q_{out} \tag{4}$$

$$V\frac{d\left[HCO_{3}^{-}\right]}{dt} = \left[HCO_{3in}^{-}\right]q_{in} - \left[HCO_{3}^{-}\right]q_{out} \qquad (5)$$

The electroneutrality equation of the chemical reaction inside the mixing tank is:

$$\left[\left(C_5 H_{12} O N^+ \right)_n \right] + \left[A l^{3+} \right] + \left[F e^{3+} \right] + \left[C a^{2+} \right] + \left[H^+ \right] = \left[H C O_3^- \right] + \left[O H^- \right]$$
(6)

The difference of the ionic concentrations can be expressed as [Gardia & Godoy, 2011]:

$$X = [HCO_{3}^{-}] - [(C_{5}H_{12}ON^{+})_{n}] - [Al^{3+}] - [Fe^{3+}] - [Ca^{2+}]$$
(7)

where

$$X = \begin{bmatrix} H^+ \end{bmatrix} - \begin{bmatrix} OH^- \end{bmatrix} \tag{8}$$

By solving the quadratic expression in (9), the $[H^+]$ values of the effluent stream flowing out of the tank can be obtained.

$$\left[H^{+}\right]^{2} - X\left[H^{+}\right] - k_{w} = 0 \tag{9}$$

The pH of the effluent from the mixing tank is expressed as:

$$pH = -\log\left[H^+\right] \tag{10}$$

The material balance expression for the mixing tank can be written as:

$$V\frac{dX}{dt} = \left[HCO_{3\ in}^{-}\right]q_{in} - \left\{\left[\left(\left(C_{5}H_{12}ON^{+}\right)_{n}\right)_{in}\right] + \left[Al_{in}^{3+}\right]\right\}q_{a} - \left[Fe_{in}^{3+}\right]q_{b} - \left[Ca_{in}^{2+}\right]q_{c} - Xq_{out}$$
(11)

The expression for the surface charge (SC) of the raw water is obtained as:

$$\sigma = \left[\left(\frac{2}{\pi}\right) n\epsilon\kappa T \right]^{\frac{1}{2}} \sinh 1.15 \left(pH_0 - pH \right) \tag{12}$$

Where $[(C_5H_{12}ON^+)_n]$ is the polyamine ionic concentration at the mixing tank outlet, $[((C_5H_{12}ON^+)_n)_{in}]$ the polyamine ionic concentration at the mixing tank inlet, $[Al^{3+}]$ is the aluminium ionic concentration at the mixing tank outlet, $[Al_{in}^{3+}]$ aluminium ionic concentration at the mixing tank inlet, $[Fe^{3+}]$ is the ferric ionic concentration at the mixing tank outlet, $[Fe^{3+}_{in}]$ ferric ionic concentration at the mixing tank inlet, $[Ca^{2+}]$ calcium ionic concentration at the mixing tank outlet, $[Ca^{2+}_{in}]$ calcium ionic concentration at the mixing tank outlet, $[Ca^{2+}_{in}]$ $[HCO_3^-]$ bicarbonate ionic concentration of the effluent stream, $[HCO_{3in}^-]$ bicarbonate ionic concentration of influent stream, q_a flow rate of sudfloc 3835 solution, q_b flow rate of ferric chloride solution, q_c flow rate of hydrated lime, q_{out} flow rate of the effluent stream, q_{in} flow rate of the influent stream, V volume, $[H^+]$ hydrogen ions concentration, $[OH^-]$ hydroxide ions concentration, k_w dissociation constant of water, σ surface charge SC, κ Boltzman constant, T temperature, ϵ relative dielectric permittivity, pH_o pH at point of zero charge and n ionic strength.

The dynamics of the coagulation chemical dosing unit can be described using (10), (11), and (12).

The states of the model are:

$$x = [C_1 \ C_2 \ C_3 \ C_4 \ C_5]^T \tag{13}$$

The controlled variables or measured outputs are:

$$y = [SC \ pH]^T \tag{14}$$

The manipulated variables or control inputs are:

$$u = \left[q_a \ q_b \ q_c\right]^T \tag{15}$$

where C_1 concentration of the $((C_5H_{12}ON^+)_n + Al^{3+})$ ions, C_2 concentration of (Fe^{3+}) ions, C_3 concentration of (Ca^{2+}) ions, C_4 concentration of (HCO_3^-) ions, C_5 difference between the concentration of hydrogen (H^+) and hydroxide (OH^-) ions, SC surface charge of the effluent water, and pH pH of the effluent water.

3.2 Process Identification

The system identification problem dealt with in this study is to estimate the dynamic model of the coagulation chemical dosing unit using nonlinear autoregressive external input (NLARX) technique. NLARX is a discrete time models employed for system identification of nonlinear systems. The general expression is:

$$y(t) = F[y(t-1), \dots, y(t-n_y), u(t-1), \dots, u(t-n_u), \epsilon(t-1), \dots, \epsilon(t-n_{\epsilon})] + \epsilon(t)$$
(16)

where F is a nonlinear function and ϵ prediction errors.

Assuming the input data is sufficient to excite the system persistently. The problem is formulated as finding the non linear function that relates explicitly the exciting input signals with the sampled outputs of the system.

For the identification of the dosing unit, a set of three different excitation signals were presented to the nonlinear model of the dosing unit. These excitation signals are sampled time-domain data at a sampling rate of 60 seconds. The input data presented to the model is shown in Fig. 2 and the output data obtained from the model is shown in Fig. 3. Table 1 shows the parameters for the simulation of the process model.

The black-box modelling approach was applied to obtain the nonlinear model that fit into the input-output dataset. All variables were normalised and have comparable numerical ranges. Using this approach, the selection of the



Fig. 2. Input data of the model



Fig. 3. Output data of the model

nonlinearity estimators and model order was done through a set of experimental trials to obtained a satisfactory result [Ljung et al, 2007]. The nonlinearity estimators examined and applied for the NLARX model were: sigmoid network; wavelet network; and tree partition.

The order and delay matrices of the model structure were computed using the delayed input and output variables knows as regressors. These are generally represented as:

$$y(t-1), y(t-2), \dots, y(t-n_a), u(t),$$

 $u(t-1), \dots, u(t-n_b-1)$ (17)

where n_a is $(n_y \times n_y)$ matrix indicating the past output terms used to predict the current output (y), n_b is $(n_y \times n_u)$ matrix indicating past input terms used to predict the current output (y) and n_k is the $(n_y \times n_u)$ matrix showing the delay from the inputs to the outputs in terms of the number of samples. n_u and n_y represent



Fig. 4. Comparison of the nonlinear estimators of the model

the three inputs and two outputs of the process model respectively.

The best result of the experiments is shown in Fig. 4. It is observed that wavenet estimator had the best fit to the measured output data among the three estimators. The input, output order and delay matrices of the model are:

$$n_a = \begin{bmatrix} 2 & 2 \\ 2 & 2 \end{bmatrix} \qquad n_b = \begin{bmatrix} 1 & 1 & 1 \\ 1 & 1 & 1 \end{bmatrix} \qquad n_k = \begin{bmatrix} 1 & 1 & 1 \\ 1 & 1 & 1 \end{bmatrix}$$

The nonlinear model obtained was linearised at an operating point to give a first-order Taylor series approximation. The operating point for the model was $[-2.09 \times 10^{-4} \, \mu eq/mg \ 7.9 \, pHunit]$. The linearised model approximation in form of state space model is described in the Appendix.

3.3 Model Predictive Control (MPC)

MPC is a control strategy for predicting the future response of a plant at each control interval by computing a sequence of control variable manipulations for any system that its model can be developed. Fig. 5 shows the block diagram of MPC where the control law generates a control sequence that drives the future behaviour of the system to be equal to the setpoint values. The key objective of the MPC is to find the input sequence that minimises the cost function or performance criterion of a system over a prediction horizon based on a desired output trajectory. The cost function operates on the deviations between the predicted model output and the reference trajectory, and the change in the input (input sequence) over the control horizon.

In Fig. 5, y(k+i), $i = 1, \ldots, p$ represents the process outputs over a future time interval (prediction horizon, p), r(t+i) is set point values, u(k) is first control move, $\varepsilon(k)$ is prediction error, $y_m(k)$ is model prediction and



Fig. 5. Block diagram of model predictive control

y(k) is the plant measurement. MPC generates a number of possible control signals, and select the control signals which give the prediction that minimise errors from the reference trajectories over the time interval. This results into an optimisation problem that is solved to effect the model predictive control action at time k [Galvez-Carrillo et al., 2009, Bemporad et al., 2013]:

$$J = \underbrace{\min_{C}}_{C} \{ \sum_{i=0}^{p-1} \sum_{i=1}^{n_{y}} |w_{i+1,j}^{y} (y_{j} (k+i+1|k) - r_{j} (k+i+1)|^{2} + \sum_{j=1}^{n_{u}} |w_{i,j}^{\Delta u} \Delta u_{j} (k+i|k)|^{2} + \sum_{j=1}^{n_{u}} |w_{i,j}^{u} (u_{j} (k+i|k) - u_{j,target} (k+i))|^{2} + \rho_{\varepsilon} \varepsilon^{2} \}$$

$$(18)$$

subject to the following constraints:

$$u_{j,min}(i) - \epsilon V_{j,min}^{u}(i) \le u_{j}(k+1|k) \le u_{j,max}(i) + \epsilon V_{j,max}^{u}(i)$$
(19)

$$\Delta u_{j,min}\left(i\right) - \epsilon V_{j,min}^{\Delta u}\left(i\right) \le \Delta u_{j}\left(k+1|k\right) \le \Delta u_{j,max}\left(i\right) + \epsilon V_{j,max}^{u}\left(i\right) \qquad (20)$$

$$y_{j,min}\left(i\right) - \epsilon V_{j,min}^{y}\left(i\right) \le y_{j}\left(k+1|k\right) \le y_{i\,max}\left(i\right) + \epsilon V_{i\,max}^{u}\left(i\right)$$

$$(21)$$

$$\Delta u \left(k + h | k \right) = 0 \tag{22}$$

$$\epsilon \ge 0 \tag{23}$$

where $C = \Delta u (k|k), \ldots, \Delta u (m-1+k|k), \epsilon, i = 0, \ldots, p-1$, $h = m, \ldots, p-1$, p prediction horizon, m control horizon, n_y number of output variables, n_u number of input variables, $r_j (k + i + 1)$ is j^{th} component of reference vector for time $k+i+1, y_j (k+i+1|k)$ is j^{th} component of output vector predicted for time $k+i+1, \Delta u_j (k+i+1|k)$ is j^{th} component of input sequence vector predicted for time $k + i, u_j (k+i|k)$ is j^{th} component of input vector predicted for time $k + i, \epsilon$ is the slack variable or constraint softening, $w_{i,j}^{\Delta u}$ is nonnegative weight for the input increment, $w_{i,j}^{u}$ nonnegative weight for the output variables, $w_{i+1,i}^{y}$ nonnegative weight for the output variables, $u_{j,min}$

Table 1. Process modelling variables

Variable and Symbols	Values and units
Sudfloc 3835 ions concentration,	
$\left(\left[\left(C_5H_{12}ON^+\right)_n\right] + \left[Al^{3+}\right]\right)$	$0.0001~{\rm mol/L}$
Ferric ions	
concentration, $\left[Fe^{3+}\right]$	$0.0001~{\rm mol/L}$
Calcium ions	
concentration, $\left[Ca^{2+}\right]$	0.0001 mol/L
Bicarbonate ion	
concentration, $\left[HCO_3^{-}\right]$	$0.00001~{\rm mol/L}$
Hydrogen ion	
concentration, $\left[H^+\right]$	10^{-7}
Tank volume, V	29,000 Litres
Dissociation constant	
of water, k_w	10^{-14}
Temperature, T	298 K
Ionic strength, n	$50 \ge 10^6 \text{ mol/L}$
Relative dielectric permittivity, ϵ	80
Boltzman constant, κ	$1.38 \ge 10^{23} J K^{-1}$

lower bound of component of the input vector, $u_{j,max}$ upper bound of component of the input vector, $\Delta u_{j,min}$ lower bound of component of the input increment vector, $\Delta u_{j,max}$ upper bound of component of the input increment vector, $y_{j,max}$ upper bound of component of the output vector, $y_{j,max}$ upper bound of component of the output vector, $u_{j,target} (k+i)$ a setpoint for the component of the input vector for time k + i, ρ_{ϵ} slack variable weight, $V_{j,min}^{u}$, $V_{j,max}^{u}$ equal concern for the relaxation of input vector constraints, $V_{j,min}^{\Delta u}$, $V_{j,max}^{\Delta u}$ equal concern for the relaxation of input sequence vector constraints and $V_{j,min}^{y}$, $V_{j,max}^{y}$ equal concern for the relaxation of output vectors constraints.

4. SIMULATION RESULTS AND DISCUSSIONS

The NMPC was simulated for three different scenarios for a period of 24 hours. A time delay of 1 second was assumed for the sensors required for the output variables measurement. The sampling time for the controller was 60 seconds. Table 2 shows the parameters specified for the NMPC controller. However, the weights on the input variables were specified as zero whereas no constraint was specified for the output variables to allow them move freely. The three scenarios are presented as follows:

Scenario one: In this simulation condition, the performance of the controller to track changes in set points every six hours was examined. The reference trajectories were multi-step signal between -0.00014 and -0.00012 $\mu eq/mg$ for the surface charges and between 7.5 and 8.5 pH unit for the pH of the effluent stream. The performance of the NMPC simulation is shown in Fig. 6. The result shows the ability of NMPC to track the reference trajectories set for the two output variables satisfactorily.

Scenario two: Here, the performance of NMPC was compared with that of linearised and unconstrained version of NMPC controller (LTIMPC). Fig. 7 illustrates the comparison of the two controllers and their performances on the dosing unit. It can be observed from Fig. 7 that the performances of controllers are the same. The values of integral of squared error (ISE) used as the performance metric for the two controller was 1048400. It indicates



Fig. 6. Response of the NMPC to changes in setpoints

 Table 2. Parameters specification of the model

 Predictive Control

Parameters	Values
Prediction horizon, p	15
Control horizon, m	4
Constraint: Sudfloc 3835 flow	$0 < q_a < 2$
Constraint: Ferric chloride flow	$0 < q_b < 2$
Constraint: Hydrated lime	$0 < q_c < 2$
Weight: Surface charge	1
Weight: pH	1
Rate Weight for input variables	0.2

that NMPC compares favourably with LTIMPC. It is not inferior in performance to the LTIMPC that behaves like linear model predictive control.

Scenario three: The response of the controller under disturbances was evaluated in this simulation test. The flowrate q_a was set as the manipulated variable, q_b and q_c as the measured and unmeasured disturbance inputs respectively. The input disturbances were modelled as step signals. In addition, a randomly generated noise signal was introduced to the manipulated input. Fig. 8 shows the performance of the NMPC. The controller performed satisfactory by rejecting the disturbances applied to the dosing unit. Fig. 9 shows the control moves of the NMPC. This simulation demonstrates the ability of the NMPC as a good feedfoward-feedback controller and its robustness to noise signals and perturbations.

5. CONCLUSIONS

This study considers the setpoint tracking and disturbance rejection performance of the nonlinear MPC when applied to a coagulation chemical dosing unit of water treatment plants. Simulation tests of the control system are examined under different conditions. The study shows that the NMPC based control strategy is adequate and effective to maintain the output variables at constant level and reject disturbances introduced to the chemical dosing unit. The



Fig. 7. Comparison of the performances of NMPC and LTIMPC



Fig. 8. Performance of NMPC under disturbance rejection and robustness test

satisfactory performance exhibited by NMPC makes it a suitable controller for water coagulation process where the traditional control approaches degrade with time. Further work will include applications of neural network, fuzzy and multiple model predictive controllers to the the dosing unit and evaluation of their performances.

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Fig. 9. Control moves of NMPC

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Appendix A. LINEARISED MODEL OF THE COAGULATION CHEMICAL DOSING UNIT

$$\Delta x (k+1) = A' \Delta x (k) + B' \Delta u (k)$$

$$\Delta y (k) = C' \Delta x (k) + D' \Delta u (k)$$
(A.1)

where_

 $a_{21} = -19.6; a_{22} = 0.99; a_{23} = -2.1 \times 10^{-4}; a_{24} = -8.74 \times 10^{-4}; a_{25} = -7.1 \times 10^{-5}$

$$B' = \begin{bmatrix} 0 & 0 & 0 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 1 \end{bmatrix}$$
$$C' = \begin{bmatrix} c_{11} & c_{12} & c_{13} & c_{114} & c_{15} \\ c_{21} & c_{22} & c_{23} & c_{24} & c_{25} \end{bmatrix}$$
$$c_{11} = 0.1; \ c_{12} = -1.42 \times 10^{-9}; \ c_{13} = -1.93 \times 10^{-9}; \ c_{14} = 1.36 \times 10^{-8}; \ d_{15} = 1.6 \times 10^{-9}$$

 $c_{21} = -19.6; c_{22} = 0.99; c_{23} = -2.1 \times 10^{-4}; c_{24} = -8.74 \times 10^{-4}; c_{25} = -7.1 \times 10^{-5}$