



**PARAMETER AND STATE ESTIMATION IN
CHROMATOGRAPHIC SMB PROCESSES
WITH INDIVIDUAL COLUMNS AND
NONLINEAR ADSORPTION ISOTHERMS**

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Abstract:

In this paper, measurement based parameter and state estimation in Simulated Moving Bed plants with nonuniform columns is investigated. The estimation strategy presented uses the available measurements of the concentrations in the product flows and in one internal flow which is realistic for industrial applications. The estimation task is solved in a decentralized fashion. The correction of the parameters and the state is performed only for the column positioned in front of the respective measurement. Convergence is achieved by the shift of the product concentration measurements. The local estimation problems are solved by Extended Kalman filters. The scheme is validated for a propranolol isomers system with nonlinear adsorption isotherms.

Keywords: Simulated Moving Bed chromatography, parameter estimation, state estimation, Extended Kalman filter, decentralized estimation

1. INTRODUCTION

Preparative chromatographic separation processes are an established separation technology in downstream processing in the pharmaceutical and fine chemicals industries. Most industrial applications are performed discontinuously, leading to low productivity and high solvent consumption. In recent years, continuous Simulated Moving Bed SMB processes are increasingly applied due to their advantages with respect to the utilization of the adsorbent and reduced solvent consumption. The SMB process consists of several chromatographic columns which are interconnected in series to con-

stitute a closed loop. An effective counter current movement of the liquid phase and the solid phase is achieved by periodical and simultaneous switching of the inlet and the outlet ports by one column in the direction of the liquid flow (Figure 1).

Since SMB processes are characterized by mixed discrete and continuous dynamics, spatially distributed state variables with steep slopes, and slow and strongly nonlinear responses of the concentrations profiles to changes of the operating parameters, they are difficult to control. An overview of recent achievements in the optimization and control of chromatographic separations can be found in (Engell and Toumi, 2005). In (Toumi and Engell, 2004a) and (Toumi and Engell, 2004b), a nonlinear optimizing control scheme was proposed and successfully applied to a 3-zone reac-

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tive SMB process for glucose isomerization. In each switching period, the operating parameters are optimized to minimize a cost function. The product purities appear as constraints in the optimization problem. In the optimization, a rigorous model of the general rate type is used. Plant/model mismatch is taken into account by error feedback of the predicted and the measured purities. In addition, the model parameters are regularly updated. In (Toumi *et al.*, 2005), the control concept was extended to the more complex processes Varicol and Powerfeed that offer a larger number of degrees of freedom that can be used for the optimization of the process economics while satisfying the required product purities. A slightly different approach to the control of SMB processes was reported by (Erdem *et al.*, 2004a) and (Erdem *et al.*, 2004b). Here, the online optimization is based upon a linearized reduced model which is corrected by a Kalman filter that uses the concentration measurements in the product streams. In this work, the switching period is considered as fixed, while in the previously mentioned work it is a parameter in the optimization. In (Toumi and Engell, 2004a) and (Toumi and Engell, 2004b), the prediction is based on the assumption that the columns are uniform (i.e. they all show the same behavior) and that the modelling errors are small. However, the properties of each individual column differ since they have different effective lengths, different packings with adsorbent and catalyst (for the case of reactive chromatography) and the column temperatures can exhibit some variation. In this paper, an estimation concept is presented for the estimation of parameters and states of chromatographic columns with individual properties. We assume that measurements of the concentrations in one internal and in both product streams are available. The estimation of the column parameters can be used to detect degradations of a column during continuous operation of the plant.

The remainder of this paper is structured as follows: in the next section, the model of the SMB process is introduced. Section 3 reports the observer design for the SMB plant. Simulation results are presented in section 4. Finally, a summary and outlook for future research are given.

2. PROCESS MODEL

The columns of the SMB process can be divided into four different zones according to their relative position with respect to the inlet and the outlet ports as depicted in Figure 1:

- (i) Zone I between solvent and extract port: desorption of the more strongly retained component

- (ii) Zone II between extract and feed port: desorption of the less retained component
- (iii) Zone III between feed and raffinate port: adsorption of the more strongly retained component
- (iv) Zone IV between raffinate and solvent port: adsorption of the less retained component.

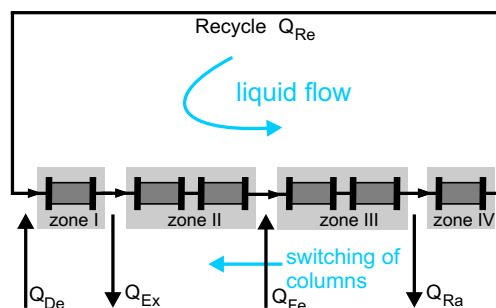


Fig. 1. Schematic diagram of the SMB process

From mass and concentration balances, the relations at the inlet and the outlet nodes can be expressed as

$$\begin{aligned}
 \text{Desorbent node : } & Q_{IV} + Q_{De} = Q_I \\
 & c_{i,IV}^{out} Q_{IV} = c_{i,I}^{in} Q_I \\
 \text{Extract node : } & Q_I - Q_{Ex} = Q_{II} \quad (1) \\
 \text{Feed node : } & Q_{II} + Q_{Fe} = Q_{III} \\
 & c_{i,II}^{out} Q_{II} + c_{i,Fe} Q_{Fe} = c_{i,III}^{in} Q_{III} \\
 \text{Raffinate node : } & Q_{Ra} + Q_{IV} = Q_{III} \\
 & i = A, B,
 \end{aligned}$$

where $Q_{I,II,III,IV}$ denote the internal flow rates of the corresponding zones I, II, III, IV, Q_{De} , Q_{Ex} , Q_{Fe} , and Q_{Ra} are the external flow rates of the respective inlet/outlet ports and, $c_{i,j}^{out}$ and $c_{i,j}^{in}$ denote the concentrations of the component i in the stream leaving or entering the respective zone j . In this paper, the separation of a racemic mixture of propranolol, a β -blocker, (Toumi *et al.*, 2003) at high purities with a 1/2/2/1 column configuration is investigated. Accurate dynamic models of multi-column continuous chromatographic processes consist of dynamic process models of the single chromatographic columns, the node balances (1) which describe the connection of the columns, and the port switching. The chromatographic columns are described accurately by the *general rate model* (Guichon *et al.*, 1994) which accounts for all important effects of a radially homogeneous column, i.e. mass transfer between the liquid and the solid phase, pore diffusion, and axial dispersion. The concentration of component i is given by c_i in the liquid phase and q_i in the solid phase. D_{ax} is the axial dispersion coefficient, u the interstitial velocity, ϵ_b the void fraction of the bulk phase, $k_{l,i}$ the film mass transfer resistance, and D_p the diffusion coefficient within the particle pores. The concentration within the pores is denoted by $c_{p,i}$. The following set of partial

differential equations can be obtained from a mass balance around an infinitely small cross-section of the column:

$$\frac{\delta c_i}{\delta t} + \left(\frac{1 - \epsilon_b}{\epsilon_b} \right) \frac{3k_{l,i}}{r_p} (c_i - c_{p,i}|_{r=r_p}) = D_{ax,i} \frac{\delta^2 c_i}{\delta z^2} - u \frac{\delta c_i}{\delta z} \quad (2)$$

$$(1 - \epsilon_b) \frac{\delta q_i}{\delta t} + \epsilon_p \frac{\delta c_{p,i}}{\delta t} - \epsilon_p D_{p,i} \frac{1}{r^2} \frac{\delta}{\delta r} \left(r^2 \frac{\delta c_{p,i}}{\delta r} \right) = 0 \quad (3)$$

with appropriate initial and boundary conditions

$$c_{i,t=0} = c_i^{in}; \quad c_{p,i,t=0} = c_{p,i}(0, r, x),$$

$$\left. \frac{\delta c_i}{\delta z} \right|_{z=0} = \frac{u}{D_{ax,i}} (c_i - c_i^{in}); \quad \left. \frac{\delta c_i}{\delta x} \right|_{z=L} = 0 \quad (4)$$

$$\left. \frac{\delta c_p}{\delta r} \right|_{r=0} = 0; \quad \left. \frac{\delta c_p}{\delta r} \right|_{r=r_p} = \frac{k_i}{\epsilon_p D_{p,i}} (c_i - c_{p,i,r=r_p}).$$

The adsorption equilibrium behavior and the system parameters for the propranolol isomers investigated here have been determined experimentally by (Toumi *et al.*, 2003). The adsorptive behavior is modelled by a modified competitive Langmuir adsorption isotherm (the components are referred to as *A* and *B*):

$$q_i = H_i^1 c_{p,i} + \frac{H_i^2 c_{p,i}}{1 + \sum_j k_j^2 c_{p,j}} \quad i = A, B. \quad (5)$$

For numerical simulation, an efficient discretization (Gu, 1995) is used where a finite element discretization of the bulk phase is combined with orthogonal collocation of the solid phase.

3. STATE AND PARAMETER ESTIMATION

3.1 Estimation strategy

A concept for parameter and state estimation of a SMB process has to take into account the available measurement information as well as the dynamics of the SMB model. We assume here that both concentrations of the species are continuously measured in the two product streams and in the recycle stream. This is the maximum amount of information that is available in a production plant. Thus, the positions of the measurements in the considered six column SMB plant vary within a cycle of operation as indicated by Figure 2. The recycle measurement is permanently located behind the last physical column while the two product measurements move with the product ports in the direction of the liquid flow by one column when a period has passed. Hence, there permanently is a measurement behind the last physical column over the whole cycle, while each of the remaining columns have a product measurement located at their respective outlet for two

periods in each cycle (one cycle has six periods). Concerning the dynamics of the SMB model, it

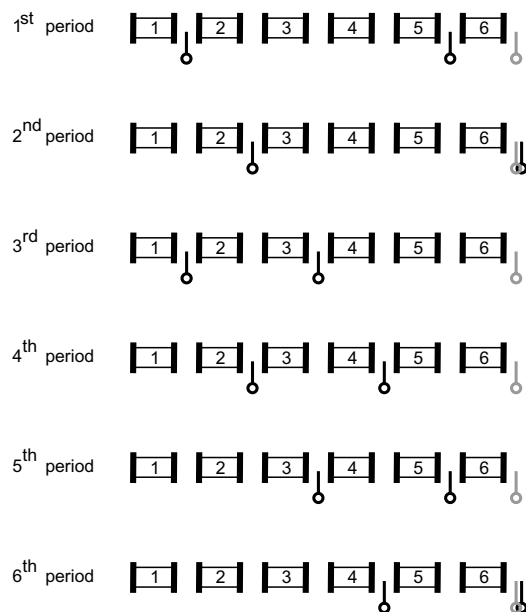


Fig. 2. Measurement positions at the physical columns for a cycle of operation (black: product streams; grey: recycle stream)

can be shown that local column parameters have a greater influence on the concentrations at the outlet of the local column compared to column parameters at a distance to the considered measurement. The influence of distant parameters is subject to a considerable time delay because the liquid flow is the link between the columns and reaches the considered column outlet only after several switching periods.

We therefore propose to perform the estimation by a set of individual, local observers that estimate the states as well as the parameters for one column only, as illustrated by Figure 3. A local observer is activated within the estimation scheme when there is a measurement located at the outlet of its respective column. The columns are of course coupled since a column with an error prone set of parameters that is in front of a column with an active local observer causes a disturbed input flow to the estimated column (indicated by the disturbance *d* in Figure 3). However, the influence of the disturbed input on the local estimation is reduced by the movement of the product measurements. One period before a local estimator is activated by a product measurement, its corresponding input is corrected by the same measurement sensor that is positioned before the column at that time. Since SMB processes are operated at a periodic steady state with high requirements for the product purities, the dissolved components move with the liquid flow by about one column within one period. Hence, the outlet concentrations of the column are influenced to a large extent by

the inlet concentrations that were measured one period earlier. Therefore, the influence of model errors of the remaining columns on the local estimation of one column is not a dominant factor in the local estimation. However, the estimated input concentration profile of column 6 for which the estimation is performed continuously deviates from the true profile and hence in this column errors of the parameter estimation are induced until a sensor is located in front of this column (see Figure 2).

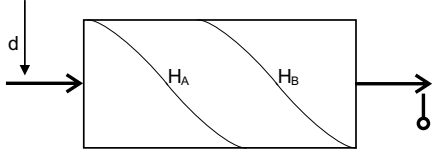


Fig. 3. Column-by-column estimation of parameters and states

3.2 Extended Kalman filter

In the local Extended Kalman filters, the parameters that are estimated are defined as states. At each time step k the dynamics \hat{f}^i of the column model are linearized around the local state estimate \hat{x}^i (each column has 100 states). The algorithm of the Extended Kalman filters is

- (1) Prediction:

$$\hat{x}_{k+1,k}^i = \hat{x}_{k,k}^i + \int_{t_k}^{t_{k+1}} \hat{f}^i(\hat{x}^i, \hat{x}^{i-1}) dt \quad (6)$$

$$P_{k+1,k}^i = A_k^i P_{k,k}^i A_k^{iT} + Q^i \quad (7)$$

- (2) Correction:

$$K_k^i = P_{k,k-1}^i C^{iT} (C^i P_{k,k-1}^i C^{iT} + R)^{-1} \quad (8)$$

$$\hat{x}_{k,k}^i = \hat{x}_{k,k-1}^i + K_k^i (y_k^i - \hat{y}_{k,k-1}^i) s_{period}^i \quad (9)$$

$$P_{k,k}^i = (I - K_k^i C^i) P_{k,k-1}^i, \quad (10)$$

where P^i is the error covariance, K^i the Kalman gain, C^i the output matrix, Q^i the state noise covariance matrix, R the measurement error covariance matrix, y^i are the measurements, and A_k^i the linearized local dynamics of column i . The EKF were tuned by varying the diagonal values of the matrices P_0 , R , and Q^i . s_{period}^i is a binary integer variable that takes the value of one if the local observer of the corresponding column is activated and zero otherwise. The activation trigger s_{period}^i of the local observers is related to Figure 2.

4. RESULTS

For the results presented, the measurements are assumed to be subject to uniformly distributed white noise with a maximum deviation of 5% of

the highest concentration value of the SMB profile. Furthermore, it is assumed that the product measurement devices are placed in front of the product pumps, otherwise a deformation of the concentration profiles would be encountered. The Henry coefficients H_i^1 are chosen as estimated parameters since they have the strongest influence on the process performance. The chosen operating point achieves high purities but is not optimal with respect to solvent consumption.

Figure 5 shows the estimation of the parameters of column 1 in the case of a step in the Henry coefficient H_A^1 (more strongly adsorbed component) of column 3 (scenario 1). The estimator of column 3 converges to the true value. The estimators of columns 1, 2, 4, and 5 are not affected by the error in column 3. However, the input of column 6 is not corrected during one period and the Henry coefficient H_A^1 of column 6 is therefore increased. When the estimator of column 3 has converged, the estimator of column 6 converges as well. Figure 6 depicts the estimation of the Henry coefficients of all six physical columns in the presence of considerable model errors (scenario 2). The individual model coefficients are initialized at the nominal values that are given in the appendix while the true values of the individual Henry coefficients of the respective columns differ considerably from the nominal values. The proposed estimation concept manages to estimate all individual Henry coefficients and to reduce the state estimation error at the beginning of a period

$$J_0 = \sqrt{\sum_{i=1}^{n_{state}} (\hat{x}(i)_{smb,0} - x(i)_{smb,0})^2} \quad (11)$$

substantially (see Figure 4), apart from the error caused by the measurement noise. In Figure 7 the

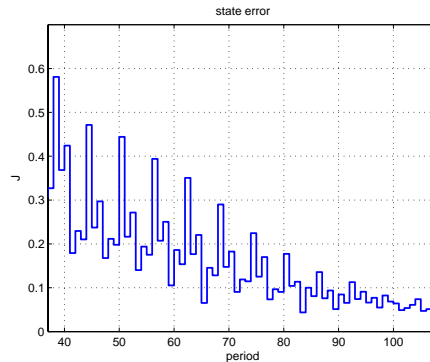


Fig. 4. State error J for scenario 2

estimation of the parameters of column 6 whose local estimator is activated over the whole cycle is compared to the corresponding measurement information for one cycle. For the investigated operating point, the concentration fronts (concentration increases or decreases drastically) are in the 2nd, 4th, and 6th period. The measured

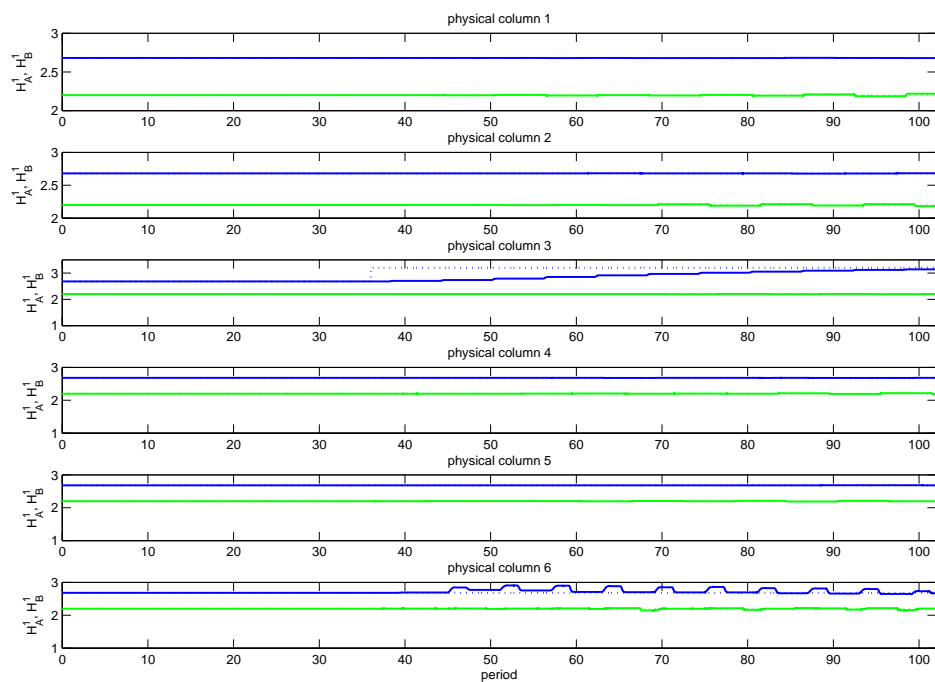


Fig. 5. Parameter perturbation of column 3 (step introduced and estimation started at the 37th period, scenario 1); lines: model, dotted lines: reference plant, black: H_A^1 , grey: H_B^1

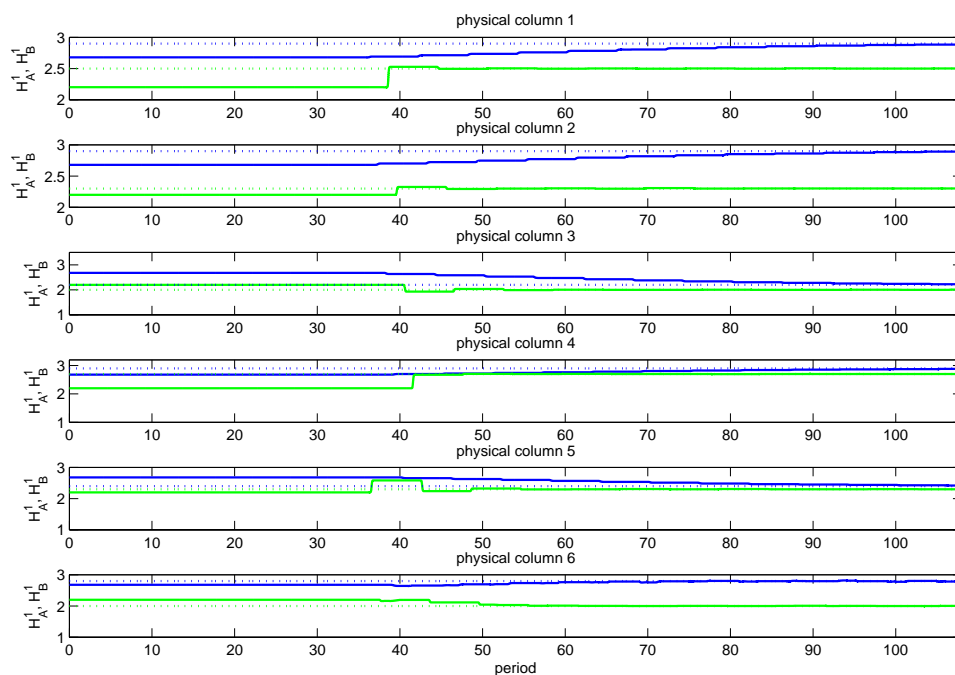


Fig. 6. Parameter estimation for wrong Henry coefficients of all columns (estimation started from the 37th period on, scenario 2); lines: model, dotted lines: reference plant, black: H_A^1 , grey: H_B^1

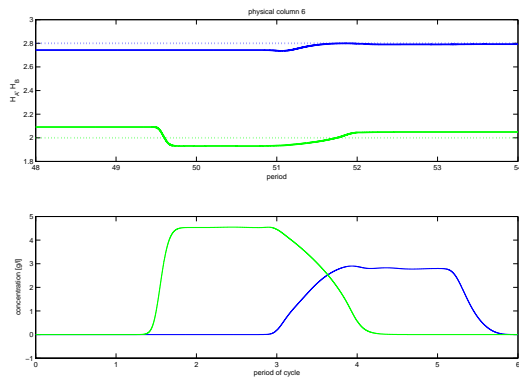


Fig. 7. Column 6 (permanent measurement): parameter estimation and measured concentration profile over one cycle for scenario 2

extract and raffinate profiles correspond to period 2 and 5 modified slightly by the individual column properties. When a concentration front moves over the extract port, the Henry coefficient H_B^1 of the respective column is corrected by the local estimation. However, at the raffinate port (period 5) only a small part of the front is measured. The correction of the Henry coefficient H_A^1 related to the more strongly adsorbed component A is rather slow. This is due to the specific operating point.

5. CONCLUSION

A parameter and state estimation scheme for an SMB process with individual columns applying local Extended Kalman filters based on only three measurement positions has been presented. The observer performs well. The individual column parameters can be reconstructed. It is expected that the performance of a model predictive control scheme can be improved by applying a decentralized state estimation. In future research, the implementation of Moving Horizon Estimator is planned.

SYSTEM PARAMETERS

separator length	$L = 10\text{cm}$
separator diameter	$D = 1\text{cm}$
adsorption coefficients	$H_A^1 = 2.68$ $H_A^2 = 0.9412$ $k_A^2 = 340 \frac{\text{cm}^3}{\text{g}}$ $k_B^2 = 262 \frac{\text{cm}^3}{\text{g}}$
film transfer resistance	$H_B^1 = 2.2$ $H_B^2 = 0.4153$ $k_{l,A} = 0.5610 \cdot 10^{-2} \frac{\text{cm}}{\text{s}}$ $k_{l,B} = 0.3310 \cdot 10^{-2} \frac{\text{cm}}{\text{s}}$
void fraction	$\epsilon_b = 0.4$
particle void fraction	$\epsilon_p = 0.5$
particle diameter	$d_p = 20\mu\text{m}$
particle diffusion coefficient	$D_p = 10^{-5} \frac{\text{cm}^2}{\text{s}}$

density	$\rho = 1.0 \frac{\text{g}}{\text{ml}}$
viscosity	$\eta = 6.8510 \cdot 10^{-4} \frac{\text{g}}{\text{cm s}}$
axial diffusion coefficient	$D_{a,x} = 10^{-6} \frac{\text{cm}^2}{\text{s}}$
feed	$Q_{Fe} = 0.31 \frac{\text{ml}}{\text{min}}$ $c_{A,Fe} = c_{B,Fe} = 7.5 \frac{\text{g}}{\text{l}}$
period	$\tau = 2.05\text{min}$
extract	$Q_{Ex} = 1.94 \frac{\text{ml}}{\text{min}}$
raffinate	$Q_{Rex} = 1.12 \frac{\text{ml}}{\text{min}}$
eluent	$Q_{De} = 2.75 \frac{\text{ml}}{\text{min}}$
recycle	$Q_{Re} = 4.80 \frac{\text{ml}}{\text{min}}$
measurement error covariance	$R = 0.01I$ (I : unity matrix)
initial error covariance	$P_0 = 300I, P_{0,6} = 30$
state noise covariance	$Q_x = 100I, Q_{x,6} = 0.01I$ $q_p = 0.05, 0.01$

ACKNOWLEDGMENT

This investigation was supported by the Deutsche Forschungsgemeinschaft (DFG) under grant DFG En 152/34. This support is gratefully acknowledged.

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