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## STEADY-STATE MULTIPLICITY FOR AUTOCATALYTIC REACTIONS IN A NONIDEAL MIXING OF CSTR WITH TWO UNPREMIXED FEEDS

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**Abstract**—The necessary and sufficient conditions for multiplicity are derived for isothermal autocatalytic reactions in a nonideal mixing of CSTR with two unpremixed feeds. Cholette's model is utilized in this paper. The ratio of the fraction of the feed for A to B entering the zone of perfect mixing,  $(n_b/n_a)$ , is indicated to determine significantly the multiplicity and uniqueness region. When the  $(n_b/n_a)$  is larger (smaller) than 1, the multiplicity region decreases (increases), and the conversion in active space increases (decreases). When  $(n_b/n_a)$  is equal to 1, the multiplicity region and the conversion in active space are the same as those in the ideal mixing. If operating in a unique region in the ideal mixing, the system may show multiplicity when  $(n_b/n_a) < 1$ . On the other hand, if operating in a multiplicity region in the ideal mixing, the system may show uniqueness when  $(n_b/n_a) > 1$ .

## 1. INTRODUCTION

The presence of multiple steady states under the non-linear model and the physical process has received a considerable amount of attention in reaction engineering literature. Since van Heerden (1953) published his work, a considerable amount of research has been directed at analyzing the multiplicity of chemical reacting systems. A number of comprehensive reviews of steady-state multiplicity in various systems have been presented (Morbidelli *et al.*, 1986; Razon and Schmitz, 1987).

Among the various reactors, continuous-flow stirred tank reactors (CSTRs) have been used extensively to study the dynamic behavior of non-linear chemical systems according to bifurcation theory and singularity theory (Balakotaiah and Luss, 1983; Gray and Scott, 1983; Kay *et al.*, 1989). However, these systems have generally been based on the ideal mixing in the reactor. Thus, their results are not useful in the imperfectly mixing CSTR.

During the past decade, Lo and Cholette (1983) investigated the multiplicity of a conversion in a cascade of imperfectly mixing CSTR's. Liou and Chien (1990a) applied Cholette's model towards analyzing the maximum steady states in the two CSTRs in series. Liou and Chien (1990b) used both Cholette's model

and a simplified Four Environment model for the sake of studying the macromixing and micromixing effect on the steady-state multiplicity in a CSTR where the reaction rate is  $-\gamma_a = kC_a/(1 + KC_a)^2$ . Furthermore, Liou and Chien (1991) also employed Cholette's model in finding an exact multiplicity criteria for input multiplicity in a CSTR. However, all of the above studies concentrated on the CSTR with one feed. Recently, Li (1994) showed that additional bifurcations may occur in an ideal CSTR if an additional inflow of reactants is introduced.

Actually, the CSTR with two unpremixed feeds is more often encountered in the chemical industry than that with one feed. Hannon and Horsthemke (1987) used a coalescence-dispersion model of the CSTR to study the effect of premixed and unpremixed reactant feeds. They showed that the region of bistability is smaller for segregated feed streams than for a fully premixed feed stream. Lintz and Weber (1980, 1987a, b) showed that the conversion depends not only on the stirring rate but also on the feed stream configuration, i.e., premixed or unpremixed condition. However, the necessary and sufficient conditions for the multiplicity and uniqueness in terms of parameters were not deduced by them. Exact multiplicity criteria for autocatalytic reactions were previously published by Lin (1979, 1981). However, those criteria could only be applied in an ideal CSTR. The criteria for steady-state multiplicity coming from the literature,

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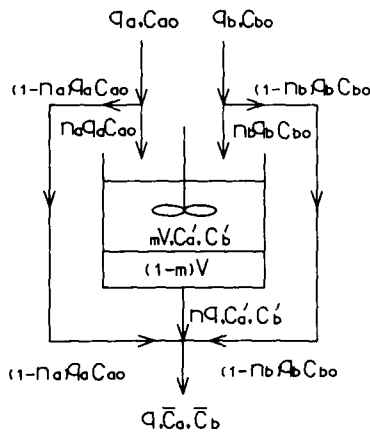


Fig. 1. Schematic representation of Cholette's model with two unpremixed feeds.

therefore, have their limitations for designing and controlling a CSTR.

The primary purposes of this paper lie in (1) establishing exact multiplicity and uniqueness criteria for an imperfectly mixing CSTR with two unpremixed feeds for autocatalytic reactions, and (2) studying the effect of by-passing and dead space on multiplicity via Cholette's model.

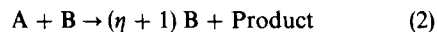
## 2. MIXING MODEL AND SYSTEM EQUATIONS

A schematic diagram of Cholette's model is illustrated in Fig. 1. The parameter  $m$  is the fraction of the total volume which is perfect mixed. The parameters  $n_a$  and  $n_b$  are the fractions of the feed  $A$  and  $B$  entering the zone of perfect mixing, respectively.

Thus, the overall fraction of the feed entering the perfect mixing system, the parameter  $n$ , can be obtained by the total mass balance in the CSTR of Fig. 1 (if density change can be ignored).

$$\frac{q_a}{q} n_a + \frac{q_b}{q} n_b = n. \quad (1)$$

An autocatalytic reaction occurring in the CSTR is considered as



with an overall rate expression given by

$$-\gamma_a = k C_a^p C_b^r \quad (p > 0, r > 0) \quad (3)$$

where  $\eta$  is the stoichiometric coefficient and  $k$  is the apparent reaction rate constant.  $p$  and  $r$  are apparent reaction orders.

The steady-state equations of the reactor are

$$n_a q_a C_{a0} - n q C'_a = m V k C_a^p C_b^r \quad (4)$$

$$n_b q_b C_{b0} + \eta (n_a q_a C_{a0} - n q C'_a) = n q C'_b \quad (5)$$

Define the conversion of  $A$  in active space,

$$Y = \frac{n_a q_a C_{a0} - n q C'_a}{n_a q_a C_{a0}}. \quad (6)$$

Both concentrations of  $A$  and  $B$  in active space can be expressed in terms of their feed concentration and conversion of  $A$ .

$$C'_a = \frac{n_a q_a}{n q} C_{a0} (1 - Y) \quad (7)$$

$$C'_b = \frac{1}{n q} (n_b q_b C_{b0} + \eta n_a q_a C_{a0} Y). \quad (8)$$

Equation (4) can be written in the following dimensionless form:

$$\frac{Y}{\bar{\theta}} = (1 - Y)^p (\bar{R} + Y)^r \quad (9)$$

where

$$\bar{\theta} = (m/n) (n_a/n)^{p+r-1} \theta \quad (10)$$

$$\theta = (kV/q) \eta^r (q_a C_{a0}/q)^{p+r-1} \quad (11)$$

$$\bar{R} = (n_b/n_a) R \quad (12)$$

and

$$R = q_b C_{b0} / \eta q_a C_{a0} \quad (13)$$

## 3. ANALYSIS OF MULTIPLE STEADY-STATES

### (a) Tangency approach

Note that eq. (9) has a form similar to that obtained by Lin (1979). Following the same procedure as stated by Lin (the details for derivation are provided in Appendix), the following necessary conditions and sufficient condition for multiplicity (uniqueness) are obtained.

### Necessary conditions for multiplicity

The necessary conditions for the system to have multiple solutions are

$$r > \bar{R} p$$

$$p + r - 1 - \bar{R} > 0$$

$$-\bar{R}(p-1) + r - 1 > 0 \quad (14)$$

$$[1 + \bar{R}(p-1) - r]^2 - 4(p+r-1)\bar{R} > 0.$$

The steady-state solution is unique for those system parameters which do not satisfy the above criteria.

### Sufficient condition for multiplicity

The sufficient condition is

$$\bar{\theta}_1 < \bar{\theta} < \bar{\theta}_2 \quad (15)$$

where

$$\bar{\theta}_1 \text{ and } \bar{\theta}_2 = \frac{\bar{Y}}{(1 - \bar{Y})^p (\bar{R} + \bar{Y})^r} \quad (16)$$

and

$$\bar{Y} = \frac{-[1 + \bar{R}(p-1) - r] \pm \{[1 + \bar{R}(p-1) - r]^2 - 4(p+r-1)\bar{R}\}^{1/2}}{2(p+r-1)}. \quad (17)$$

Necessary and sufficient conditions for the system to have multiplicity are obviously the combination of both criteria (14) and (15). The violation of the conditions (14) and (15) would guarantee the uniqueness of the system.

(b) *Singularity theory approach*

The steady-state equation of eq. (9) can be written as

$$F = (1 - Y)^p(\bar{R} + Y)^r - Y/\bar{\theta} = 0 \quad (18)$$

Following Gray and Scott (1990), since  $dF/d\bar{\theta}$  does not vanish, isola and mushroom patterns are not possible. A hysteresis loop appears in the steady-state diagram under conditions

$$F = 0 \quad \text{and} \quad \frac{dF}{dY} = 0. \quad (19)$$

Differentiating  $F$  with respect to  $Y$  yields

$$(1 - Y)^{p-1}(\bar{R} + Y)^{r-1} [r - p\bar{R} - (p + r)Y] - 1/\bar{\theta} = 0. \quad (20)$$

The necessary condition, for the solution to exist, is

$$r > p\bar{R} \quad (21)$$

Eliminating  $\bar{\theta}$  from eqs (18) and (20) one obtains

$$(p + r - 1)Y^2 + [1 + \bar{R}(p - 1) - r]Y + \bar{R} = 0 \quad (22)$$

since the two roots in eq. (22) are both between 0 and 1, their product and sum must satisfy

$$0 < \frac{\bar{R}}{p + r - 1} < 1$$

$$0 < \frac{-[1 + \bar{R}(p - 1) - r]}{p + r - 1} < 2.$$

The above conditions can be simplified as

$$p + r - 1 - \bar{R} > 0, \quad -\bar{R}(p - 1) + r - 1 > 0. \quad (23)$$

The roots of eq. (22) are

$$\bar{Y} = \frac{-[1 + \bar{R}(p - 1) - r] \pm \{[1 + \bar{R}(p - 1) - r]^2 - 4(p + r - 1)\bar{R}\}^{0.5}}{2(p + r - 1)} \quad (24)$$

since  $\bar{Y}$  must be real, the additional condition is

$$[1 + \bar{R}(p - 1) - r]^2 - 4(p + r - 1)\bar{R} > 0. \quad (25)$$

It is noted that conditions (21), (23) and (25) are exactly the same as that obtained by the tangency approach, i.e. condition (14). Therefore, the necessary conditions for multiplicity are condition (14) also.

In order to find the boundaries of  $\bar{\theta}$ , denoted as  $\bar{\theta}_1$  and  $\bar{\theta}_2$ , eq. (24) is substituted into eq. (18), and  $\bar{\theta}_1$  and  $\bar{\theta}_2$  are given by

$$\bar{\theta}_1 \text{ and } \bar{\theta}_2 = \frac{\bar{Y}}{(1 - \bar{Y})^p(\bar{R} + \bar{Y})^r} \quad (26)$$

where  $\bar{Y}$  is determined by eq. (24). Therefore, the sufficient condition for multiplicity is

$$\bar{\theta}_1 < \bar{\theta} < \bar{\theta}_2. \quad (27)$$

It is also noted that eqs (26) and (24) are exactly the same as eqs (16) and (17). Therefore, from the above analysis, the conditions (necessary and sufficient) for the system to have the multiple steady states which are developed by the tangency approach and singularity theory approach are exactly the same. However, the modern technique of singularity theory seems to be more powerful.

*Examples*

- (1) For the case of  $p = 2$  and  $r = 1$ , condition (14) does not satisfy because of  $\bar{R} > 0$ ; therefore, the steady-state solution is unique.
- (2) For the case of  $p = 1$  and  $r = 2$ , then from condition (14), the necessary condition for multiplicity is  $\bar{R} < 1/8$ . Therefore, (a) when  $\bar{R} = 1/4$ , the system is unique, (b) when  $\bar{R} = 1/9$ , then from the sufficient condition, the system has multiple steady states for  $81/32 < \bar{\theta} < 324/125$ , and the system is unique for  $\bar{\theta} < 81/32$ , or  $\bar{\theta} > 324/125$ .

#### 4. NUMERICAL SIMULATION

A system with a particular set of  $(p, r, \bar{R}, \bar{\theta})$  can be easily verified for its multiplicity or uniqueness by examining the criteria.

Figure 2 shows that the ratio  $(n_b/n_a)$  affects the multiplicity (uniqueness) region in the  $(p, r)$  plane when  $R = 0.1$ . The region above the curve indicates that the multiple steady states occur. Therefore, from Fig. 2, we see that the degree of mixing,  $(n_b/n_a)$ , has a strong effect on the multiplicity. For example, if the reaction system is  $(p, r) = (2, 2)$  i.e. at point I, the system is unique in the ideal mixing ( $n_b/n_a = 1.0$ ). However, the system has multiple steady states in the nonideal mixing when  $n_b/n_a = 0.4$ . On the other hand, if the reacting system is  $(p, r) = (1, 2)$ , i.e. at point II, it is in a multiplicity region in the ideal mixing. How-

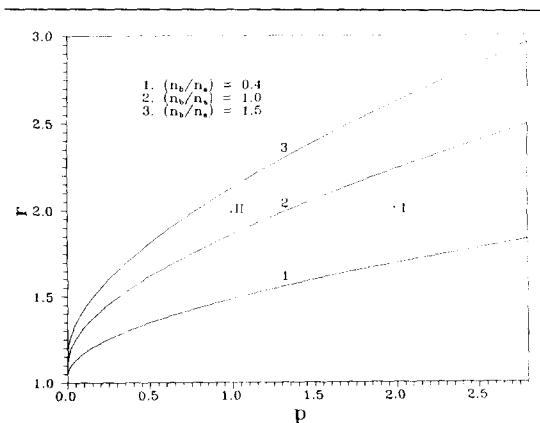


Fig. 2. The effect of  $(n_b/n_a)$  on the multiplicity region in the  $(p, r)$  plane, when  $R = 0.1$ .

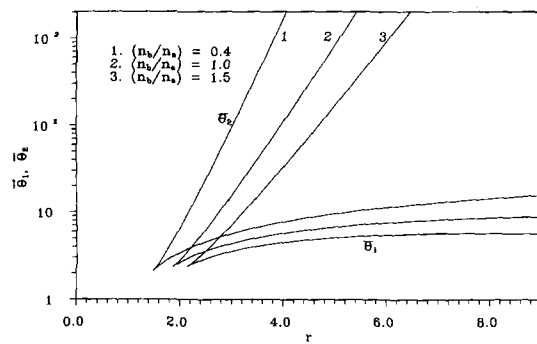


Fig. 3. The effect of  $(n_b/n_a)$  on the multiplicity region, when  $p = 1$ ,  $R = 0.1$ .

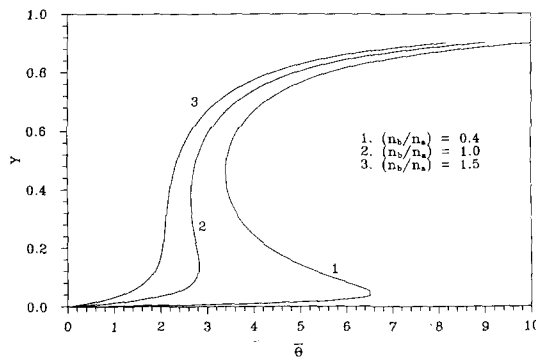


Fig. 4. The effect of  $(n_b/n_a)$  on the  $Y$ , when  $p = 1$ ,  $r = 2$  and  $R = 0.1$ .

Table 1. The necessary conditions for multiplicity with the ideal and the nonideal mixing

Ideal mixing	Nonideal mixing
$r > RP$ $p + r - 1 - R > 0$ $-R(p - 1) + r - 1 > 0$ $[1 + R(p - 1) - r]^2$ $-4(p + r - 1)R > 0$	$r > \bar{R}P$ $p + r - 1 - \bar{R} > 0$ $-\bar{R}(p - 1) + r - 1 > 0$ $[1 + \bar{R}(p - 1) - r]^2$ $-4(p + r - 1)\bar{R} > 0$

Table 2. The sufficient condition for multiplicity with the ideal and the nonideal mixing

Ideal mixing
$\theta_1 < \theta < \theta_2$
where $\theta_1 \text{ and } \theta_2 = \frac{Y}{(1 - Y)^p(R + Y)}$ $Y = \frac{-[1 + R(p - 1) - r] \pm \{[1 + R(p - 1) - r]^2 - 4(p + r - 1)R\}^{0.5}}{2(p + r - 1)}$
Nonideal mixing
$\bar{\theta}_1 < \bar{\theta} < \bar{\theta}_2$
where $\bar{\theta}_1 \text{ and } \bar{\theta}_2 = \frac{\bar{Y}}{(1 - \bar{Y})^p(\bar{R} + \bar{Y})}$ $\bar{Y} = \frac{-[1 + \bar{R}(p - 1) - r] \pm \{[1 + \bar{R}(p - 1) - r]^2 - 4(p + r - 1)\bar{R}\}^{0.5}}{2(p + r - 1)}$

ever, the system becomes unique when  $n_b/n_a = 1.5$  in the nonideal mixing.

The effect of  $\bar{\theta}$  is illustrated in Fig. 3, in which multiplicity occurs for  $R = 0.1$  and  $p = 1$ . For a given value of  $(n_b/n_a)$ , multiplicity occurs inside the region embraced by the two curves  $\bar{\theta}_1$  and  $\bar{\theta}_2$ . The solution is unique outside the region. The value of  $(n_b/n_a)$  causes the multiplicity region to shift as indicated in Fig. 3.

Figure 4 shows how  $(n_b/n_a)$  affects the conversion  $Y$  as a function of  $\bar{\theta}$  for  $p = 1$ ,  $r = 2$  and  $R = 0.1$ . When  $(n_b/n_a)$  is larger (smaller) than 1,  $Y$  is observed to become larger (smaller) than the ideal mixing. It is also noted that the parameter  $(n_b/n_a)$  strongly determines  $Y$  at the low value of  $\bar{\theta}$ , however, the effect of the  $(n_b/n_a)$  on the  $Y$  is not significant at the high value of  $\bar{\theta}$ .

Additionally, Fig. 4 shows that the multiple steady states occur for the cases of  $(n_b/n_a) = 0.4$  and 1.0, and the steady state is unique for the case of  $(n_b/n_a) = 1.5$ . This result can be identified in Fig. 2. This system is located at the point II in Fig. 2, and, as shown in Fig. 2, the point II is at the upper region for  $(n_b/n_a) = 0.4$ , and 1.0, and under the curve for  $(n_b/n_a) = 1.5$ . In Fig. 4, the tendency of multiplicity for the case of  $(n_b/n_a) = 0.4$  (multiplicity occurs at the range of  $\bar{\theta}$  from 3.41 to 6.50) is markedly more than the case of  $(n_b/n_a) = 1$  (multiplicity occurs at the range of  $\bar{\theta}$  from 2.66 to 2.88). It is fact that the curve  $(n_b/n_a = 0.4)$  is actually much lower than the curve  $(n_b/n_a = 1)$  in Fig. 2.

## 5. CONCLUSION

In general, the previous papers (Lo and Cholette, 1983; Liou and Chien, 1990a, b, 1991) which discussed mixing and multiplicity in CSTRs almost all concentrated on CSTRs with one feed. The CSTR with two unpremixed feeds is actually more often encountered in the chemical industry than that with one feed. This paper has established exact multiplicity and uniqueness criteria for an imperfectly mixing CSTR with two

unpremixed feeds for autocatalytic reactions and studied the effect of by-passing and dead space on multiplicity via Cholette's model.

The ratio of by-passing for feed A to B,  $(n_b/n_a)$ , is observed to have markedly determined the necessary and sufficient conditions. When  $(n_b/n_a)$  becomes equal to 1 ( $n_b = n_a \neq 1$ ) in the nonideal mixing, the multiplicity region and conversion in active space are the same as those obtained in the ideal mixing ( $n_b = n_a = 1$ ). When  $(n_b/n_a)$  becomes larger (smaller) than 1, the multiplicity region decreases (increases) and the conversion in active space increases (decreases). Hence, if operating in a uniqueness region in the ideal mixing, the system may consequently show multiplicity when  $(n_b/n_a) < 1$  in the nonideal mixing. On the other hand, if operating in a multiplicity region in the ideal mixing, the system may show uniqueness when  $(n_b/n_a) > 1$  in the nonideal mixing.

A comparison between the ideal and the nonideal mixing is provided in Tables 1 and 2, including the necessary and sufficient conditions for multiplicity, respectively. The observed results become significant for application in chemical reactor design once the nonideal mixing has taken place in a real reactor.

#### NOTATION

$C_{a0}, C_{b0}$	feed concentrations of A and B
$C'_a, C'_b$	concentrations of A and B in the active space
$m$	the fraction of the total volume in perfect mixing
$n_a, n_b$	the fraction of the feed entering the zone of perfect mixing for feed of A, B
$n$	the overall fraction of the feed entering the zone of perfect mixing
$p$	reaction orders of A
$R$	defined in eq. (13)
$\bar{R}$	defined in eq. (12)
$q_a, q_b$	flow rate for feed of A, B
$r$	reaction orders of B
$V$	reactor volume
$Y$	the conversion of the active space
$\bar{Y}$	critical conversions corresponding to $\bar{\theta}_1$ and $\bar{\theta}_2$

#### Greek letters

$-\gamma_a$	reaction rate per unity volume in the active space
$\eta$	stoichiometric coefficient
$\theta$	defined in eq. (11)
$\bar{\theta}$	defined in eq. (10)
$\bar{\theta}_1, \bar{\theta}_2$	defined in eq. (16)

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#### APPENDIX: THE NECESSARY CONDITION FOR MULTIPLICITY BY TANGENCY APPROACH

The steady-state equation [eq. (9)] is

$$\frac{Y}{\bar{\theta}} = (1 - Y)^p(\bar{R} + Y)^r$$

Let

$$Z_1 = \frac{Y}{\bar{\theta}} \quad (\text{A1})$$

and

$$Z_2 = (1 - Y)^p(\bar{R} + Y)^r \quad (\text{A2})$$

Steady-state conversion  $Y$  is determined from the intersection of  $Z_1(Y)$  and  $Z_2(Y)$ .

From eq. (A2), the derivative of  $Z_2$  with respect to  $Y$  is

$$\frac{dZ_2}{dY} = (1 - Y)^{p-1}(\bar{R} + Y)^{r-1}[r - \bar{R}p - Y(p + r)] \quad (\text{A3})$$

Then we see that

$$\left. \frac{dZ_2}{dY} \right|_{Y=0} > 0$$

if and only if

$$r > \bar{R}p. \quad (\text{A4})$$

Thus reaction rate for autocatalytic reaction would first increase with conversion only if  $r > \bar{R}p$ . Equation (A2) also leads to

$$\frac{dZ_2}{dY} = 0 \quad \text{at } Y = \frac{r - \bar{R}p}{p + r}. \quad (\text{A5})$$

For  $p > 0$  and  $r > \bar{R}p$ , it can be readily seen that

$$0 < \frac{r - \bar{R}p}{p + r} < 1. \quad (\text{A6})$$

Thus,  $Z_2$  has a maximum at  $Y = (r - \bar{R}p)/(p + r)$ . In addition,  $Z_2(0) > 0$  and  $Z_2(1) = 0$ . The  $Z_1$  curve is a straight line

passing through the origin with slope  $1/\bar{\theta}$ . Depending on the slope of  $Z_1$ , the two curves may have one, two or three intersections. At the tangent point, both the slopes and the ordinates of the two curves are equal. Then, we obtain

$$(p + r - 1)Y^2 + [1 + \bar{R}(p - 1) - r]Y + \bar{R} = 0. \quad (\text{A7})$$

Since the two roots in eq. (A7) are both between 0 and 1, the following conditions can be easily verified:

$$p + r - 1 - \bar{R} > 0 \quad (\text{A8})$$

$$-\bar{R}(p - 1) + r - 1 > 0. \quad (\text{A9})$$

The roots of eq. (A7) are

$$\bar{Y} = \frac{-[1 + \bar{R}(p - 1) - r] \pm \{[1 + \bar{R}(p - 1) - r]^2 - 4(p + r - 1)\bar{R}\}^{0.5}}{2(p + r - 1)}. \quad (\text{A10})$$

Since  $\bar{Y}$  must be real, the additional condition for existence of two distinct tangents from the origin is therefore

$$[1 + \bar{R}(p - 1) - r]^2 - 4(p + r - 1)\bar{R} > 0. \quad (\text{A11})$$

Thus, the necessary conditions for the system to have multiplicity solutions can be summarized in condition (14).