

## Supplemental Material:

Differential Equations describing the reaction network:

$$\frac{d(\text{aminal})}{dt} = -(k_1 + k_2 + k_3) \cdot [\text{aminal}] \quad (1)$$

$$\frac{d(OABA)}{dt} = k_3 \cdot [\text{aminal}] - k_{14} \cdot [OABA] \quad (2)$$

$$\begin{aligned} \frac{d(PABA)}{dt} &= k_2 \cdot [\text{aminal}] - k_4 \cdot [PABA]^2 - k_5 \cdot [PABA]^2 + k_8 \cdot [pPABA] \cdot [4,4' MDA] \\ &\quad - k_6 \cdot [PABA] \cdot [4,4' MDA] - k_9 \cdot [PABA] \cdot [4,4' MDA] - k_{13} \cdot [PABA] \cdot [MDA - PABA] \\ &\quad + k_7 \cdot [pPABA] + k_{10} \cdot [pPABA] \cdot [4,4' MDA] - k_{15} \cdot [PABA]^2 \end{aligned} \quad (3)$$

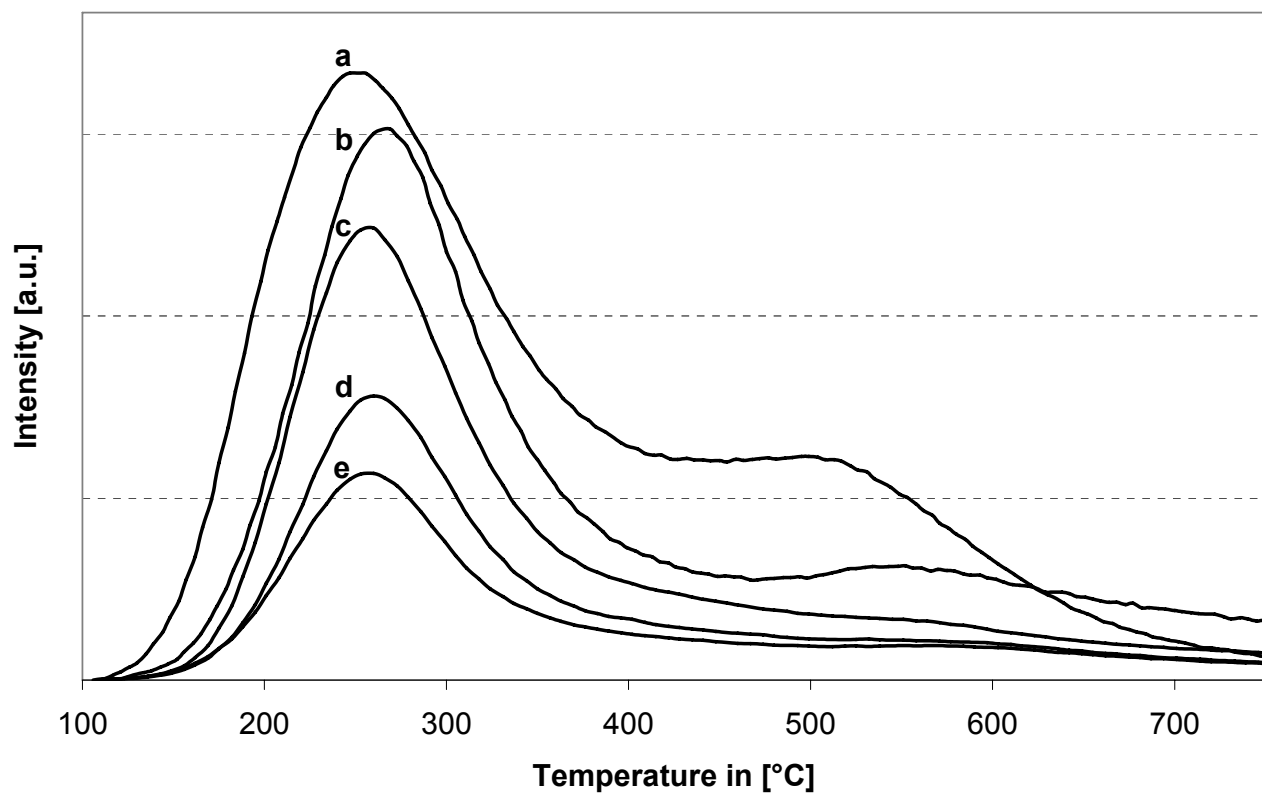
$$\begin{aligned} \frac{d(pPABA)}{dt} &= \frac{1}{2} \cdot k_2 \cdot [\text{aminal}] + \frac{1}{2} \cdot k_4 \cdot [PABA]^2 - k_7 \cdot [pPABA] - k_8 \cdot [pPABA] \cdot [4,4' MDA] \\ &\quad - k_{10} \cdot [pPABA] \cdot [4,4' MDA] + k_{13} \cdot [PABA] \cdot [MDA - PABA] \end{aligned} \quad (4)$$

$$\begin{aligned} \frac{d(MDA - PABA)}{dt} &= k_9 \cdot [PABA] \cdot [4,4' MDA] + k_{10} \cdot [pPABA] \cdot [4,4' MDA] - k_{11} \cdot [MDA - PABA] \\ &\quad - k_{12} \cdot [MDA - PABA] - k_{13} \cdot [MDA - PABA] \cdot [PABA] \end{aligned} \quad (5)$$

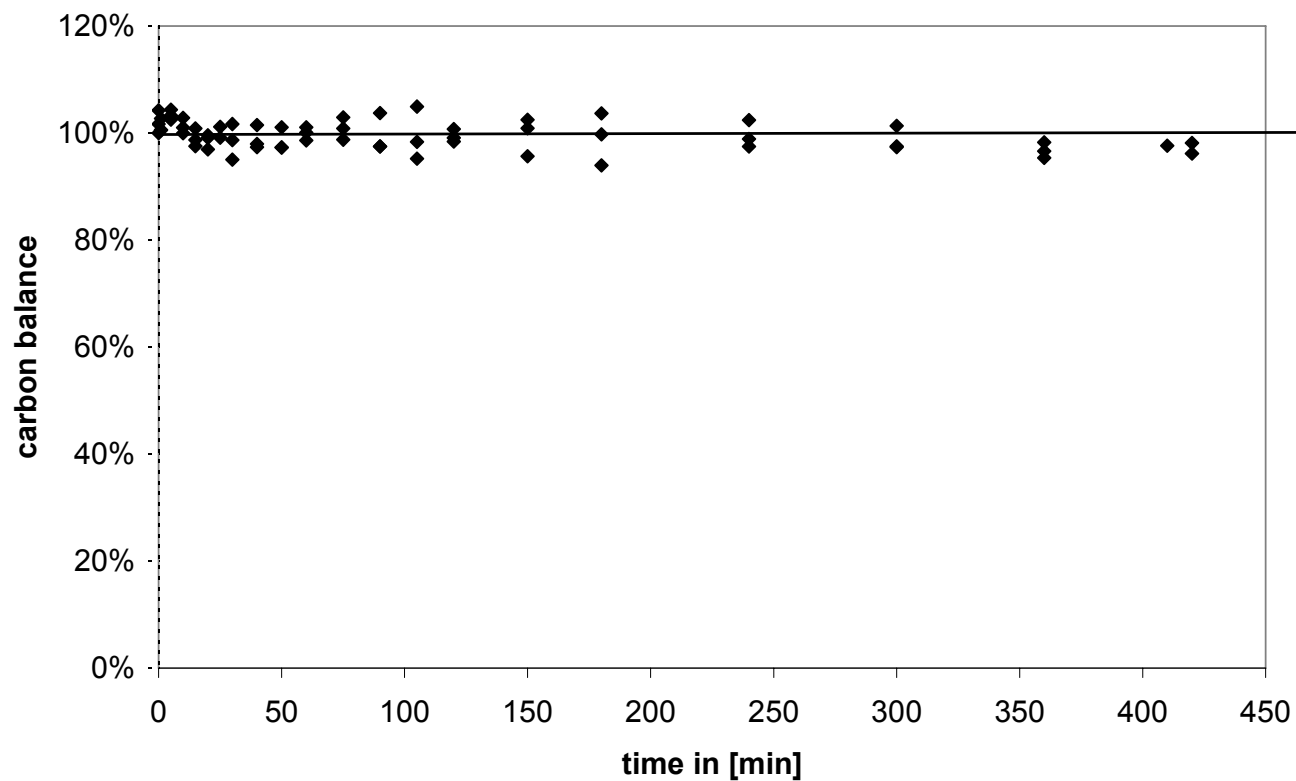
$$\begin{aligned} \frac{d(4,4' MDA)}{dt} &= k_5 \cdot [PABA]^2 + k_7 \cdot [pPABA] - k_6 \cdot [PABA] \cdot [4,4' MDA] - k_8 \cdot [pPABA] \cdot [4,4' MDA] \\ &\quad - k_9 \cdot [PABA] \cdot [4,4' MDA] - k_{10} \cdot [pPABA] \cdot [4,4' MDA] + 2 \cdot k_{12} \cdot [MDA - PABA] \\ &\quad + k_{13} \cdot [PABA] \cdot [MDA - PABA] \end{aligned} \quad (6)$$

$$\frac{d(2,4' MDA)}{dt} = k_{14} \cdot [OABA] + k_{15} \cdot [PABA]^2 \quad (7)$$

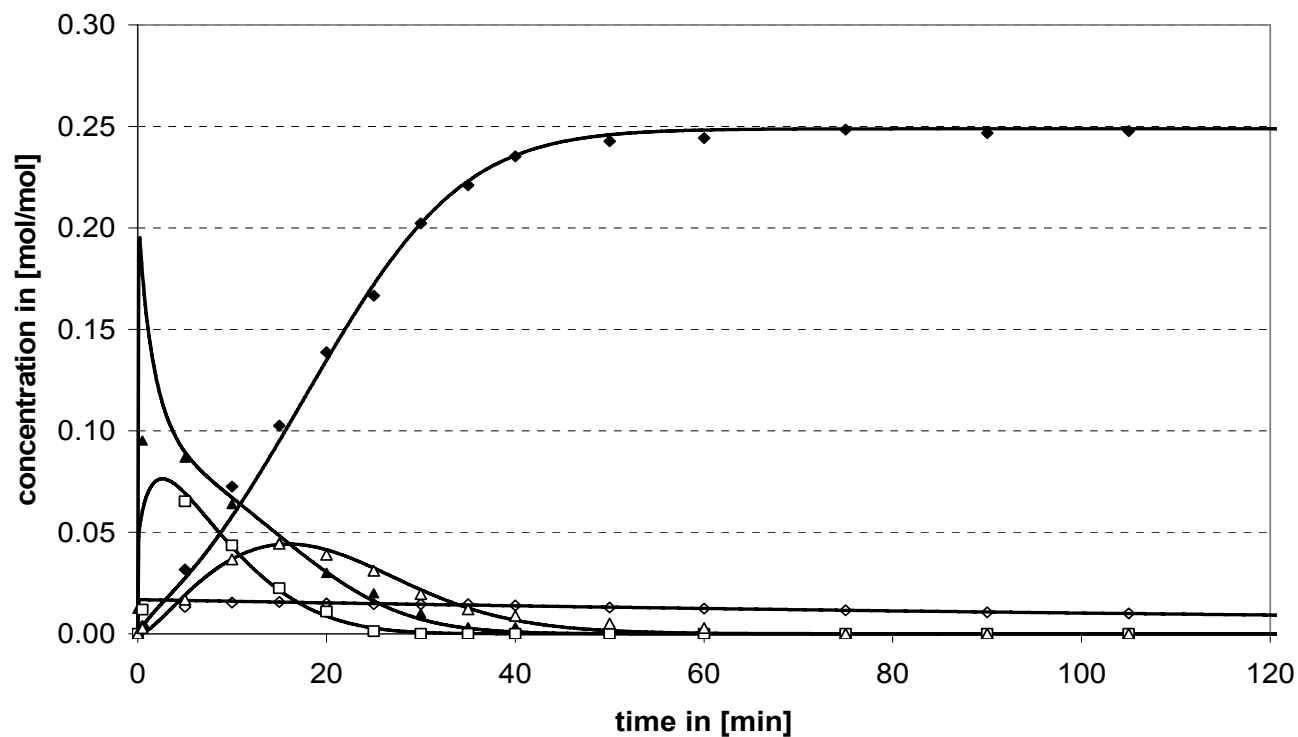
$$\frac{d(pMDA)}{dt} = k_6 \cdot [PABA] \cdot [4,4' MDA] + k_8 \cdot [pPABA] \cdot [4,4' MDA] + k_{11} \cdot [MDA - PABA] \quad (8)$$



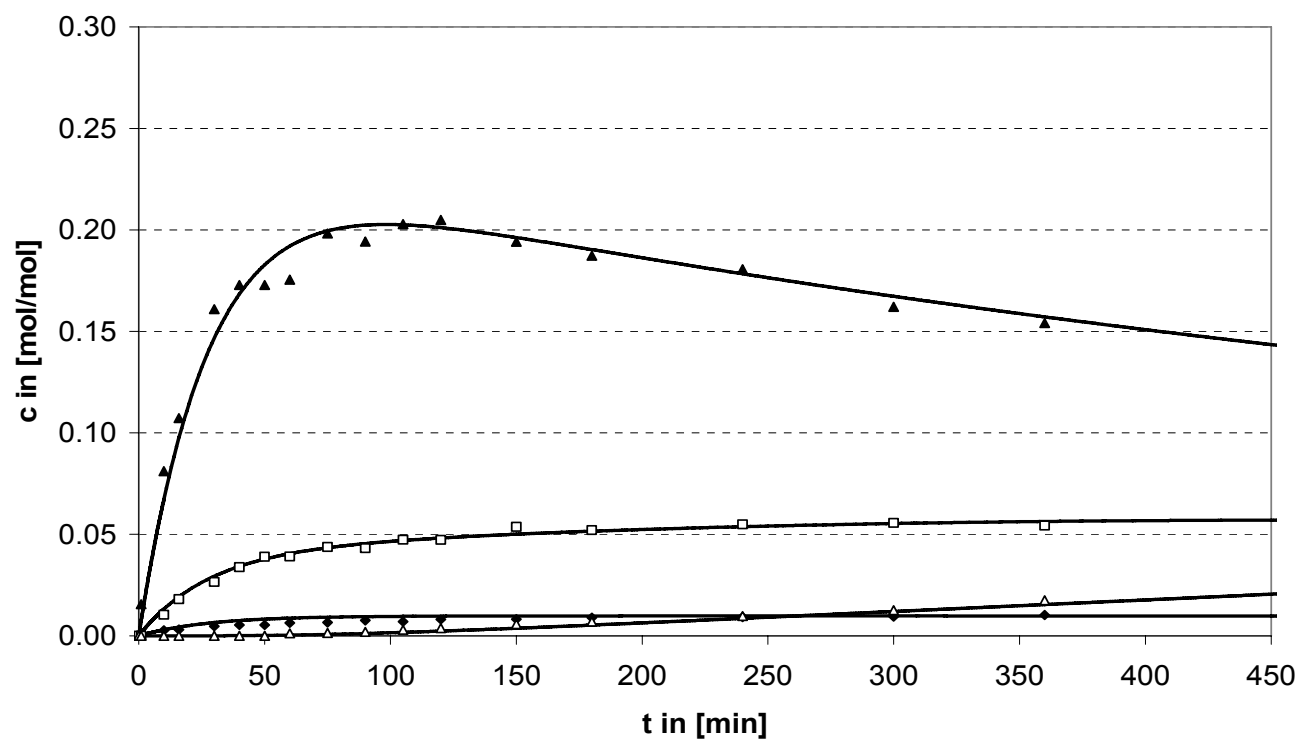
**Figure 1:** *NH<sub>3</sub>-TPD profiles of a) H-BEA 25, b) H-CBV 760, c) H/Na-CBV 760-1, d) H/Na-CBV 760-2 and e) H/Na-CBV 760-3.*



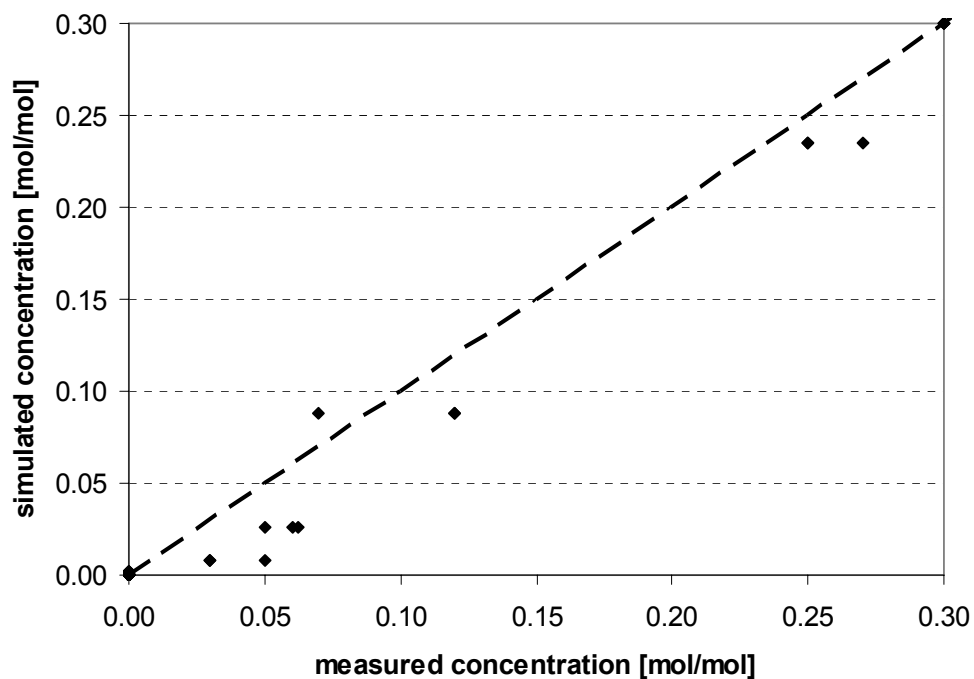
**Figure 2:** Carbon balance profile of the catalytic test reaction at 100°C over H-CBV 760.



**Figure 3:** Simulated time concentration profile (full lines) of the synthesis of MDA from aMinal over a dealuminated  $\gamma$ -type zeolite (CBV 760) at 140°C;  $\blacktriangle$  PABA,  $\diamond$  OABA,  $\blacklozenge$  4,4'-MDA,  $\square$  pPABA,  $\triangle$  MDA-PABA.

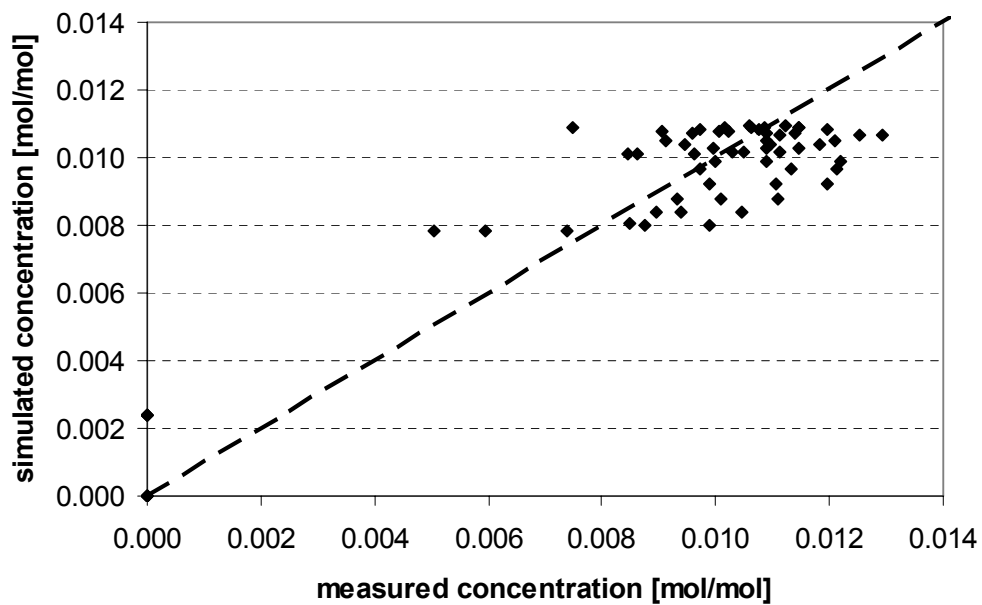


**Figure 4:** Simulated time concentration profile (full lines) of the synthesis of MDA from amination over a dealuminated  $\gamma$ -type zeolite (CBV 760) at 60°C;  $\blacktriangle$  PABA,  $\diamond$  OABA,  $\blacklozenge$  4,4'-MDA,  $\square$  pPABA,  $\triangle$  MDA-PABA.



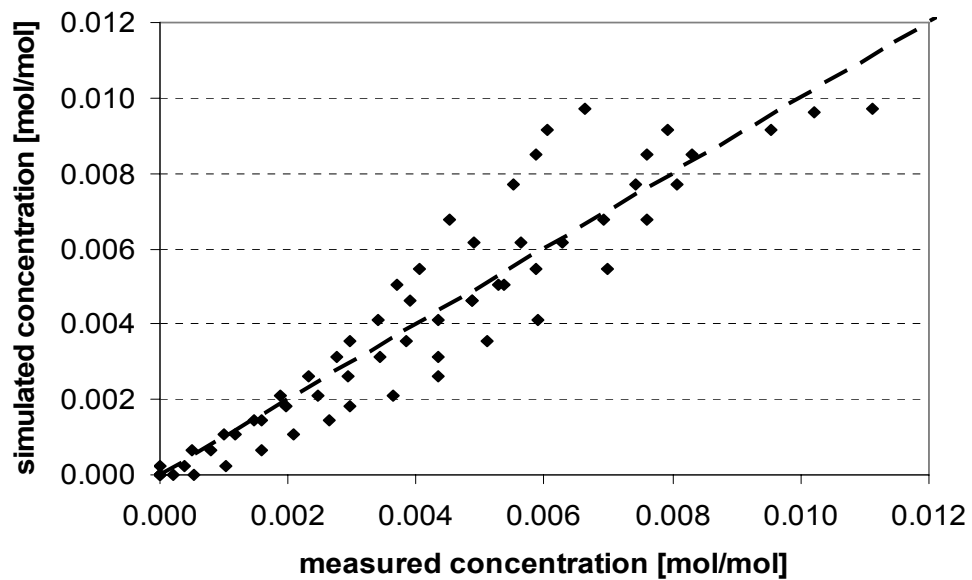
**Figure 5:** Parity plot for simulated and measured aminal concentration over H-CBV 760 at 100°C.

*Dashed line represents ideal match.*



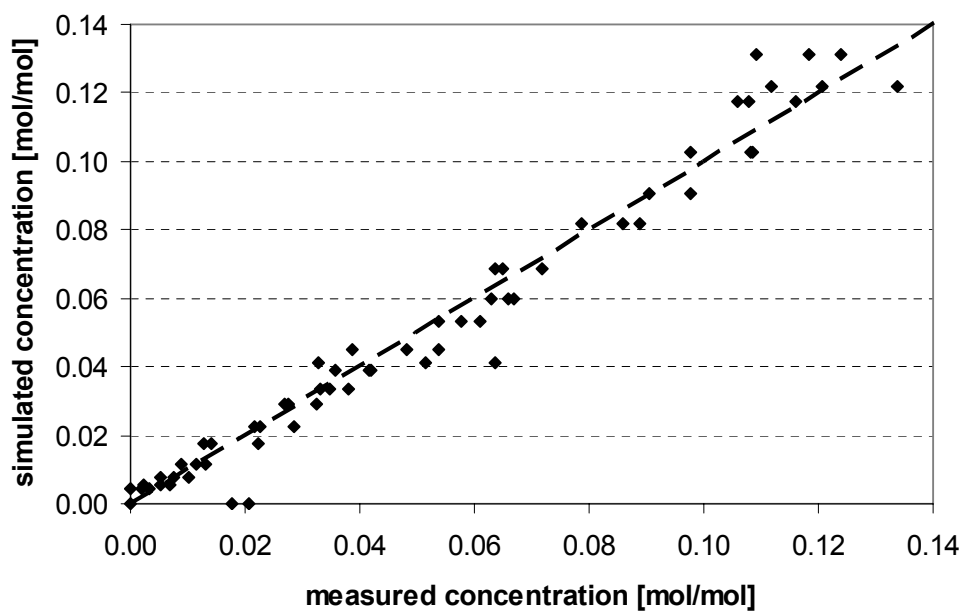
**Figure 6:** Parity plot for simulated and measured OABA concentration over H-CBV 760 at 100°C.

*Dashed line represents ideal match.*



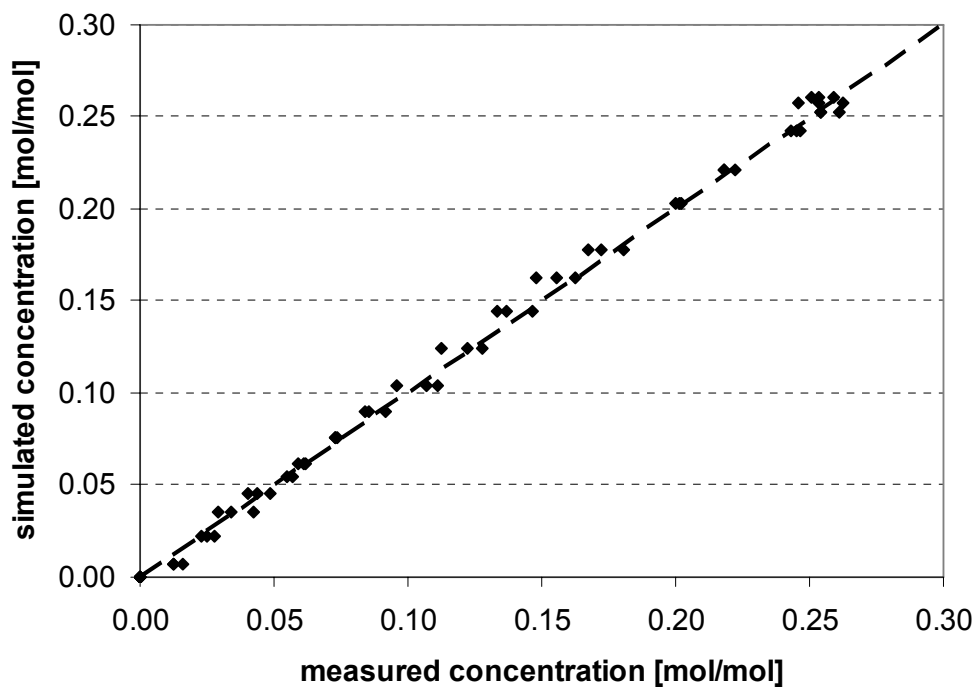
**Figure 7:** Parity plot for simulated and measured 2,4'-MDA concentration over H-CBV 760 at 100°C.

*Dashed line represents ideal match.*



**Figure 8:** Parity plot for simulated and measured PABA concentration over H-CBV 760 at 100°C.

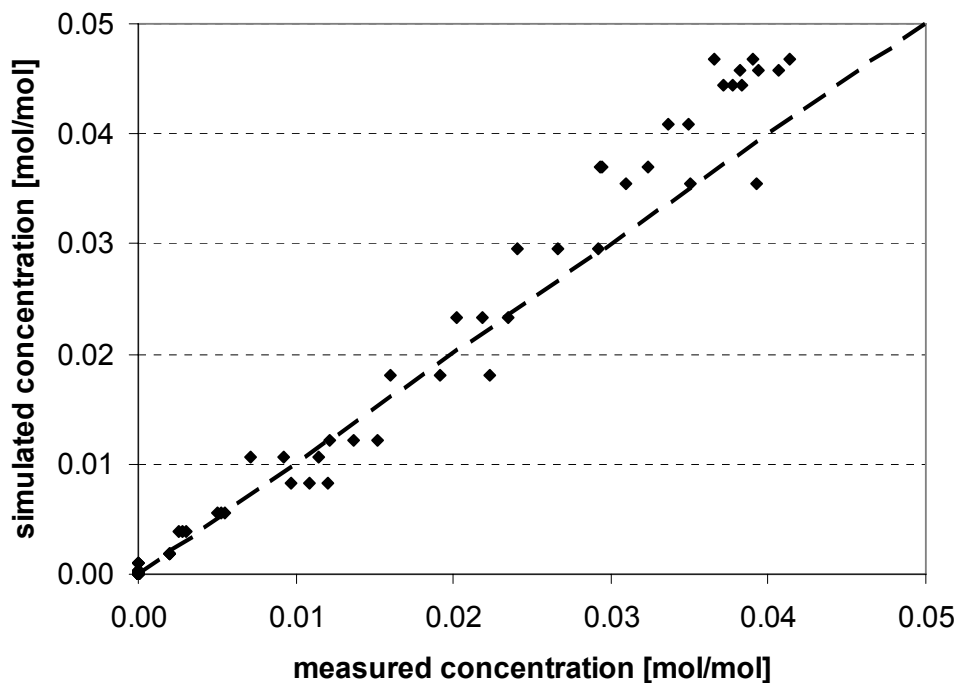
*Dashed line represents ideal match.*



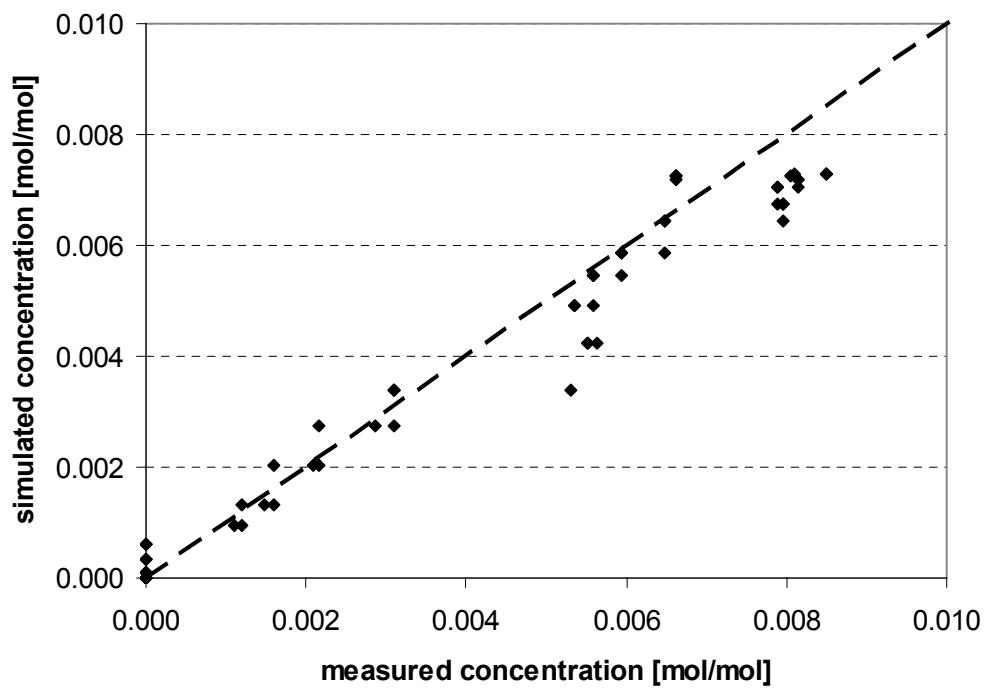
**Figure 9:** Parity plot for simulated and measured 4,4'-MDA concentration over H-CBV 760 at 100°C.

*Dashed line represents ideal match.*



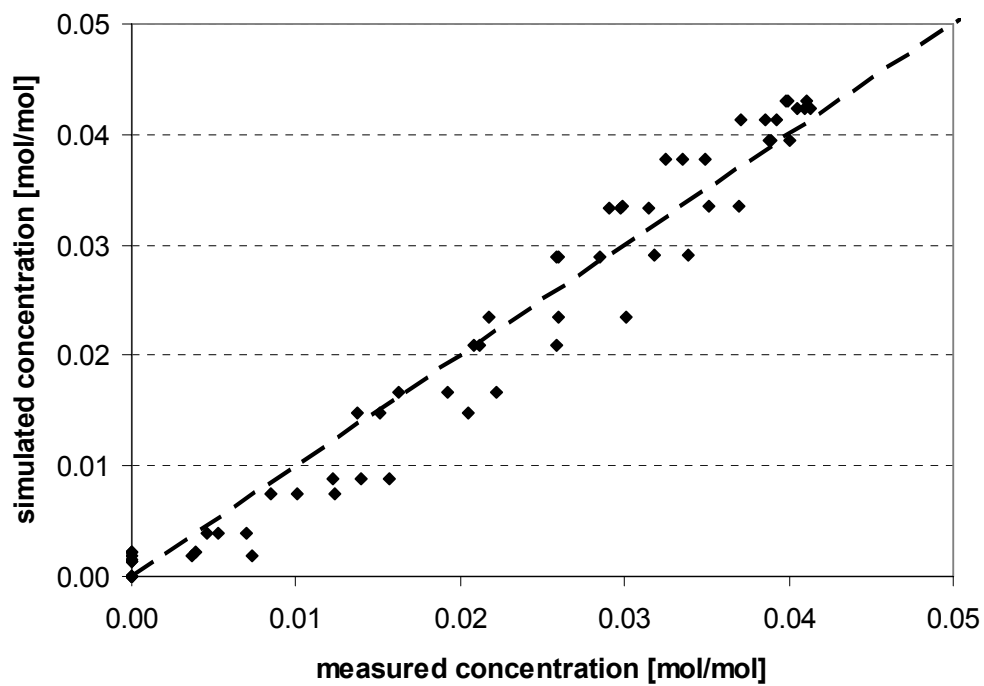


**Figure 10:** Parity plot for simulated and measured pPABA concentration over H-CBV 760 at 100°C.  
*Dashed line represents ideal match.*



**Figure 11:** Parity plot for simulated and measured pMDA concentration over H-CBV 760 at 100°C.

*Dashed line represents ideal match.*



**Figure 12:** Parity plot for simulated and measured MDA-PABA concentration over H-CBV 760 at 100°C. Dashed line represents ideal match.