

Table S1 Values of rate constants for reductive dehalogenation of bromoform and TBAA under different initial bimetallic dosages in Cu/Fe bimetallic system

| Cu/Fe  | Parameters  | Bromoform           | TBAA                 |
|--------|---|---------------------|----------------------|
| 5g/L   | $k_{\text{obs}}(\text{h}^{-1})$                   | 0.76                | 1.15                 |
|        | $k_{\text{SA}}(\text{L}/\text{h}\cdot\text{m}^2)$ | $6.8\times 10^{-2}$ | $10.3\times 10^{-2}$ |
|        | $\rho_{\text{a}}(\text{m}^2/\text{L})$            | 11.11               | 11.11                |
|        | $R^2$   | 0.945               | 0.987                |
| 7.5g/L | $k_{\text{obs}}(\text{h}^{-1})$                   | 0.99                | 1.56                 |
|        | $k_{\text{SA}}(\text{L}/\text{h}\cdot\text{m}^2)$ | $5.9\times 10^{-2}$ | $9.4\times 10^{-2}$  |
|        | $\rho_{\text{a}}(\text{m}^2/\text{L})$            | 16.67               | 16.67                |
|        | $R^2$   | 0.954               | 0.991                |
| 10g/L  | $k_{\text{obs}}(\text{h}^{-1})$                   | 1.52                | 1.84                 |
|        | $k_{\text{SA}}(\text{L}/\text{h}\cdot\text{m}^2)$ | $6.8\times 10^{-2}$ | $8.3\times 10^{-2}$  |
|        | $\rho_{\text{a}}(\text{m}^2/\text{L})$            | 22.22               | 22.22                |
|        | $R^2$   | 0.962               | 0.998                |
| 15g/L  | $k_{\text{obs}}(\text{h}^{-1})$                   | 2.02                | 2.57                 |
|        | $k_{\text{SA}}(\text{L}/\text{h}\cdot\text{m}^2)$ | $6.1\times 10^{-2}$ | $7.7\times 10^{-2}$  |
|        | $\rho_{\text{a}}(\text{m}^2/\text{L})$            | 33.33               | 33.33                |
|        | $R^2$   | 0.959               | 0.998                |
| 20 g/L | $k_{\text{obs}}(\text{h}^{-1})$                   | 2.98                | 3.34                 |
|        | $k_{\text{SA}}(\text{L}/\text{h}\cdot\text{m}^2)$ | $6.7\times 10^{-2}$ | $7.5\times 10^{-2}$  |
|        | $\rho_{\text{a}}(\text{m}^2/\text{L})$            | 44.44               | 44.44                |
|        | $R^2$   | 0.989               | 0.992                |

Table S2 Values of rate constants for reductive dehalogenation of bromoform and TBAA under different initial bimetallic dosages in Pd/Fe bimetallic system

| Pd/Fe  | Parameters  | Bromoform            | TBAA                 |
|--------|---|----------------------|----------------------|
| 5g/L   | $k_{\text{obs}}(\text{h}^{-1})$                   | 0.84                 | 0.99                 |
|        | $k_{\text{SA}}(\text{L}/\text{h}\cdot\text{m}^2)$ | $7.5\times 10^{-2}$  | $8.9\times 10^{-2}$  |
|        | $\rho_{\text{a}}(\text{m}^2/\text{L})$            | 11.11                | 11.11                |
|        | $R^2$   | 0.979                | 0.984                |
| 7.5g/L | $k_{\text{obs}}(\text{h}^{-1})$                   | 1.35                 | 1.78                 |
|        | $k_{\text{SA}}(\text{L}/\text{h}\cdot\text{m}^2)$ | $8.1\times 10^{-2}$  | $10.7\times 10^{-2}$ |
|        | $\rho_{\text{a}}(\text{m}^2/\text{L})$            | 16.67                | 16.67                |
|        | $R^2$   | 0.936                | 0.991                |
| 10g/L  | $k_{\text{obs}}(\text{h}^{-1})$                   | 1.63                 | 2.18                 |
|        | $k_{\text{SA}}(\text{L}/\text{h}\cdot\text{m}^2)$ | $7.3\times 10^{-2}$  | $9.8\times 10^{-2}$  |
|        | $\rho_{\text{a}}(\text{m}^2/\text{L})$            | 22.22                | 22.22                |
|        | $R^2$   | 0.959                | 0.999                |
| 15g/L  | $k_{\text{obs}}(\text{h}^{-1})$                   | 3.44                 | 3.31                 |
|        | $k_{\text{SA}}(\text{L}/\text{h}\cdot\text{m}^2)$ | $10.3\times 10^{-2}$ | $9.9\times 10^{-2}$  |
|        | $\rho_{\text{a}}(\text{m}^2/\text{L})$            | 33.33                | 33.33                |
|        | $R^2$   | 0.995                | 0.998                |
| 20 g/L | $k_{\text{obs}}(\text{h}^{-1})$                   | 3.75                 | 3.47                 |
|        | $k_{\text{SA}}(\text{L}/\text{h}\cdot\text{m}^2)$ | $8.4\times 10^{-2}$  | $7.8\times 10^{-2}$  |
|        | $\rho_{\text{a}}(\text{m}^2/\text{L})$            | 44.44                | 44.44                |
|        | $R^2$   | 0.998                | 0.993                |

Table S3 Values of rate constants for reductive dehalogenation of bromoform and TBAA under different pH condition in Cu/Fe bimetallic system

| Cu/Fe    | Parameters  | Bromoform            | TBAA                 |
|----------|---|----------------------|----------------------|
| Acidic   | $k_{\text{obs}}(\text{h}^{-1})$                   | 2.33                 | 2.63                 |
|          | $k_{\text{SA}}(\text{L}/\text{h}\cdot\text{m}^2)$ | $10.5\times 10^{-2}$ | $11.8\times 10^{-2}$ |
|          | $\rho_{\text{a}}(\text{m}^2/\text{L})$            | 22.22                | 22.22                |
|          | $R^2$   | 0.899                | 0.991                |
| Neutral  | $k_{\text{obs}}(\text{h}^{-1})$                   | 1.52                 | 1.84                 |
|          | $k_{\text{SA}}(\text{L}/\text{h}\cdot\text{m}^2)$ | $6.8\times 10^{-2}$  | $8.3\times 10^{-2}$  |
|          | $\rho_{\text{a}}(\text{m}^2/\text{L})$            | 22.22                | 22.22                |
|          | $R^2$   | 0.962                | 0.998                |
| Alkaline | $k_{\text{obs}}(\text{h}^{-1})$                   | 0.17                 | 0.05                 |
|          | $k_{\text{SA}}(\text{L}/\text{h}\cdot\text{m}^2)$ | $7.7\times 10^{-3}$  | $2.3\times 10^{-3}$  |
|          | $\rho_{\text{a}}(\text{m}^2/\text{L})$            | 22.22                | 22.22                |
|          | $R^2$   | 0.929                | 0.964                |

Table S4 Values of rate constants for reductive dehalogenation of bromoform and TBAA under different pH condition in Pd/Fe bimetallic system

| Pd/Fe    | Parameters  | Bromoform            | TBAA                 |
|----------|---|----------------------|----------------------|
| Acidic   | $k_{\text{obs}}(\text{h}^{-1})$                   | 2.56                 | 2.46                 |
|          | $k_{\text{SA}}(\text{L}/\text{h}\cdot\text{m}^2)$ | $11.5\times 10^{-2}$ | $11.1\times 10^{-2}$ |
|          | $\rho_{\text{a}}(\text{m}^2/\text{L})$            | 22.22                | 22.22                |
|          | $R^2$   | 0.943                | 0.986                |
| Neutral  | $k_{\text{obs}}(\text{h}^{-1})$                   | 1.63                 | 2.18                 |
|          | $k_{\text{SA}}(\text{L}/\text{h}\cdot\text{m}^2)$ | $7.3\times 10^{-2}$  | $9.8\times 10^{-2}$  |
|          | $\rho_{\text{a}}(\text{m}^2/\text{L})$            | 22.22                | 22.22                |
|          | $R^2$   | 0.959                | 0.999                |
| Alkaline | $k_{\text{obs}}(\text{h}^{-1})$                   | 0.21                 | 0.04                 |
|          | $k_{\text{SA}}(\text{L}/\text{h}\cdot\text{m}^2)$ | $9.5\times 10^{-3}$  | $1.8\times 10^{-3}$  |
|          | $\rho_{\text{a}}(\text{m}^2/\text{L})$            | 22.22                | 22.22                |
|          | $R^2$   | 0.951                | 0.975                |