Electronic supplementary information

Developing catalytic materials for the oxidative coupling of methane through statistical analysis of literature data

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Kinds of empirical models considered in our research

i. The traditional quadratic response surface model,

$$f(x,w) = w_0 + \sum_{j=1}^{d} \left(w_j x_j + w_{j,j} x_j^2 \right) + \sum_{j=2}^{d} \sum_{k=1}^{j-1} w_{j,k} x_j x_k \,. \tag{1}$$

Here, as in the main text of the paper, d is the dimension of input data x, and p is the dimension of model parameters w. If the number n of training pairs was not sufficient to learn all p parameters of the model (2), then only the purely quadratic model was learned instead:

$$f(x,w) = w_0 + \sum_{j=1}^{d} \left(w_j x_j + w_{j,j} x_j^2 \right). \tag{2}$$

ii. Clustered RBF network with diagonal Gaussian radial basis functions centered in data:

$$f(x,w) = w_{k,0} + \sum_{i=1}^{B_k} \sum_{j \epsilon D_k} w_{k,(i-1)d_k+j} e^{-\frac{\left(x_j - x_j^{(k,i)}\right)^2}{w_{k,(B_k+i-1)d_k+j}}} \; if \; x \in C_k, k = 1, \ldots, n_C, \tag{3}$$

where n_C is the number of clusters, $\mathcal{M} \subset \bigcup_{k=1}^{n_C} C_k, C_k \cap C_{k'} = \emptyset$ for $k \neq k'$,

D_k is the set of d_k dimensions to which the modeling in the cluster C_k is restricted, $\emptyset \neq D_k \subset \{1, \ldots, n\} \; for \; k = 1, \ldots, n_C$,

$w_{k,(B_k+i-1)d_k+j} > 0$ for $i = 1, \ldots, B_k, j \in D_k, k = 1, \ldots, n_C$,

B_k is the number of basis functions used for modeling in the cluster C_k

$x^{(k,1)}, \ldots, x^{(k,B_k)} \in \mathcal{M} \cap C_k$ for $k = 1, \ldots, n_C$, and $x^{(k,1)}, \ldots, x^{(k,B_k)}$ fulfill

$$E\left(w_{k,0} + \sum_{j \epsilon D_k} w_{k,j} e^{-\frac{\left(x_j - x_j^{(k,1)}\right)^2}{w_{k,d_k+j}}}, \mathcal{M} \cap C_k \right) = \min_{\xi \in \mathcal{M} \cap C_k} E\left(w_{k,0} + \sum_{j \epsilon D_k} w_{k,j} e^{-\frac{\left(x_j - \xi_j\right)^2}{w_{k,d_k+j}}}, \mathcal{M} \cap C_k \right). \tag{4}$$

If $B_k > 1$, then for $l = 1, \ldots B_k$, $\mathcal{M}_k^l = \mathcal{M} \cap C_k \setminus \left\{ x^{(k,1)}, \ldots, x^{(k,l)} \right\}$, also

$$E\left(w_{k,0}+\sum_{i=1}^{l+1}\sum_{j\epsilon D_k} w_{k,(i-1)d_k+j}e^{-\frac{\left(x_j-x_j^{(k,i)}\right)^2}{w_{k,(B_k+i-1)d_k+j}}},\mathcal{M}_{\mathrm{k}}^{\ell}\right)$$

$$=\min_{\xi\in\mathcal{M}_{\mathrm{k}}^{\ell}} E\left(w_{k,0}+\sum_{j\epsilon D_k} w_{k,ld_k+j}e^{-\frac{\left(x_j-\xi_j\right)^2}{w_{k,2ld_k+j}}}\right.$$

$$\left.+\sum_{i=1}^{l}\sum_{j\epsilon D_k} w_{k,(i-1)d_k+j}e^{-\frac{\left(x_j-x_j^{(k,i)}\right)^2}{w_{k,(B_k+i-1)d_k+j}}},\mathcal{M}_{\mathrm{k}}^{\ell}\right).$$

iii. Clustered RBF network with diagonal Gaussian radial basis functions centered in data, combined with linear regression:

$$f(x,w)=w_{k,0}+\sum_{j\epsilon D_k} w_{k,2B_kd_k+j}x_j$$

$$+\sum_{i=1}^{B_k}\sum_{j\epsilon D_k} w_{k,(i-1)d_k+j}e^{-\frac{\left(x_j-x_j^{(k,i)}\right)^2}{w_{k,(B_k+i-1)d_k+j}}}\ \text{if } x\in C_k, k=1,\dots,n_C, \tag{5}$$

where again the conditions (4) hold.

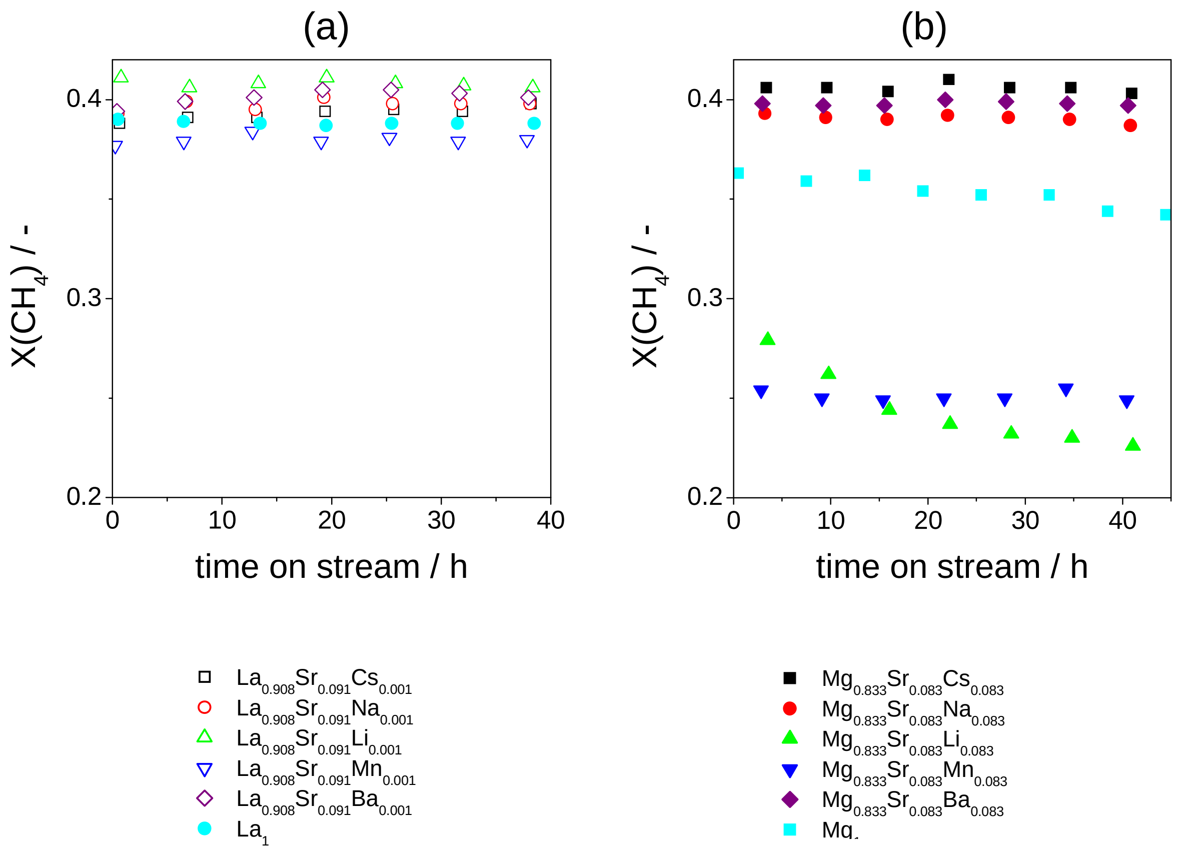

Fig. S 1 Time-on-stream methane conversion obtained over (a) La-Sr- (●-La_2O_3, □-LaSrCs, ○-LaSrNa, △-LaSrLi, ▽-LaSrMn, ◇-LaSrBa) and (b) Mg-Sr-based (■-MgO, ■-MgSrCs, ●-MgSrNa, ▲-MgSrLi, ▼-MgSrMn, ◆-MgSrBa) catalysts at 1073 K and τ of 0.0039 g min ml^{-1} using a feed of 29% CH_4 in air.

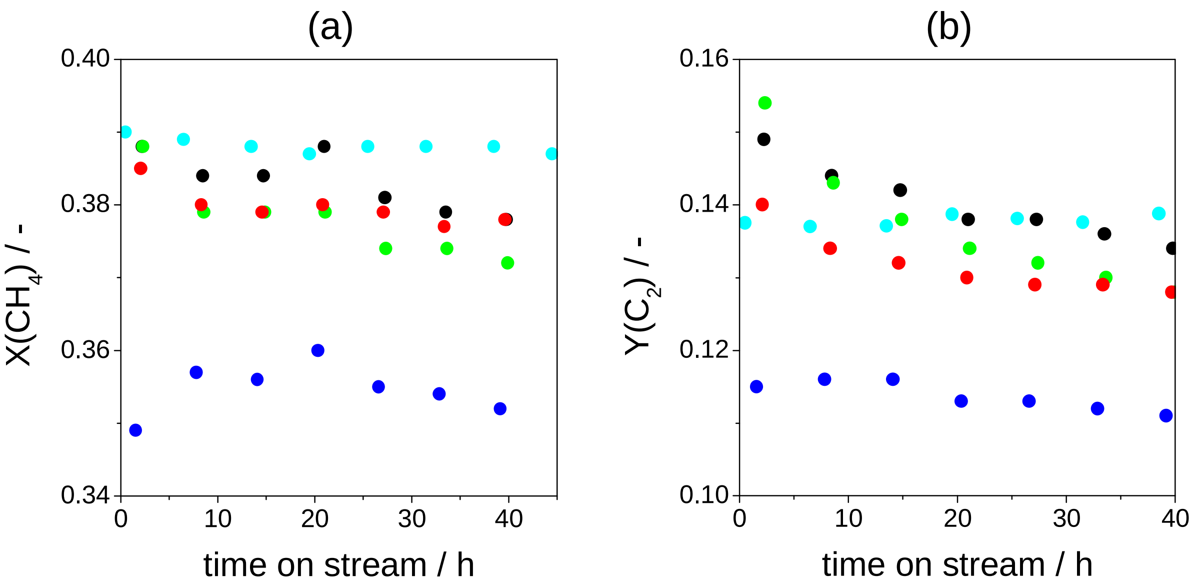

Fig. S 2 Time-on-stream (a) methane conversion and (b) yield of C_2 hydrocarbons obtained over ●-La_2O_3, ●-LaBaLi, ●-LaBaNa, ●-LaBaCs, ●-LaBaMn at 1073 K and τ of 0.0039 g min ml^{-1} using a feed of 29% CH_4 in air.

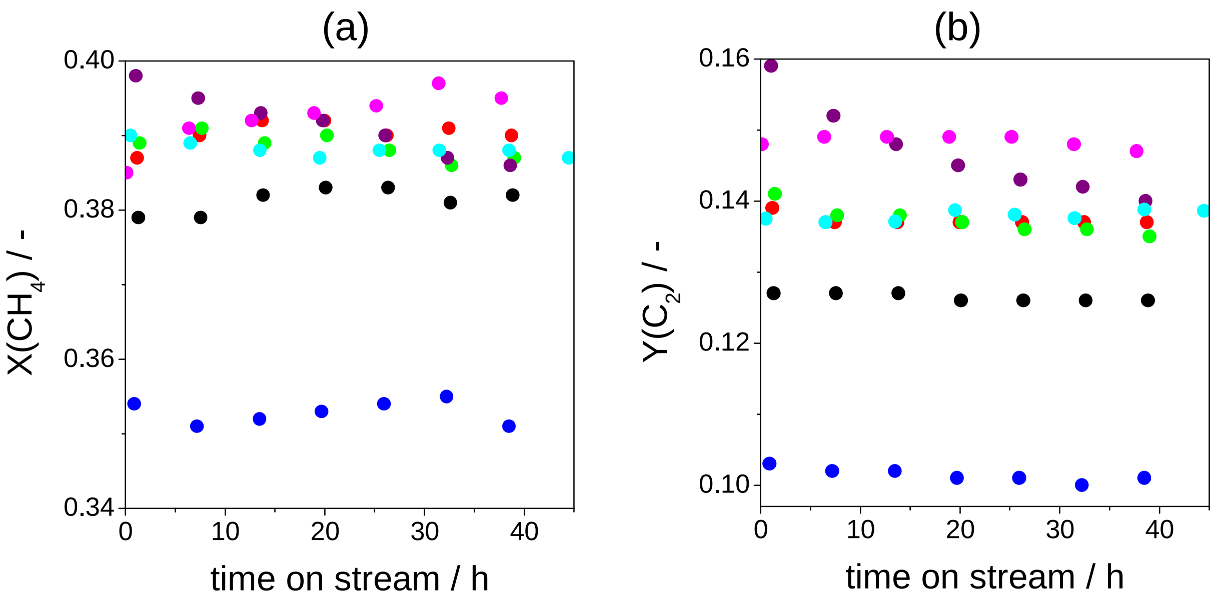

Fig. S 3 Time-on-stream (a) methane conversion and (b) yield of C_2 hydrocarbons obtained over ●-La_2O_3, ●-LaMgLi, ●-LaMgNa, ●-LaMgCs, ●-LaMgMn, ●-LaMgBa and ●-LaBaSr at 1073 K and τ of 0.0039 g min ml^{-1} using a feed of 29% CH_4 in air.

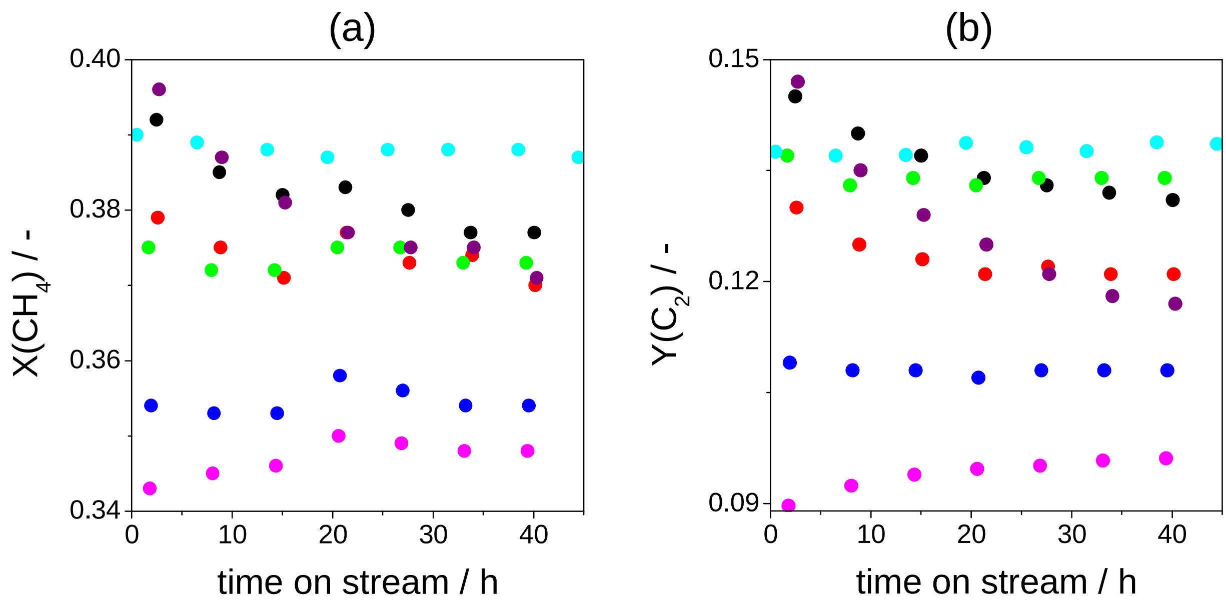

Fig. S 4 Time-on-stream (a) methane conversion and (b) yield of C_2 hydrocarbons obtained over ●-La_2O_3, ●-LaNaLi, ●-LaNaCs, ●-LaNaMn, ●-LaLiMn, ●-LaCsLi and ●-LaCsMn at 1073 K and τ of 0.0039 g min ml^{-1} using a feed of 29% CH_4 in air.

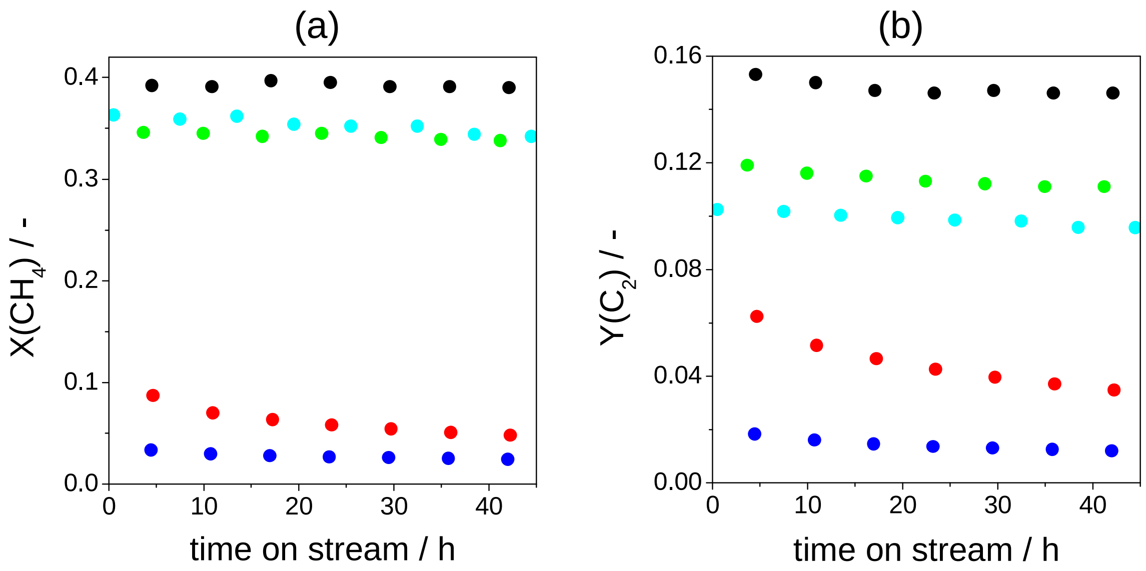

Fig. S 5 Time-on-stream (a) methane conversion and (b) yield of C_2 hydrocarbons obtained over ●-MgO, ●-MgBaLi, ●-MgBaCs, ●-LaBaNa, ●-LaBaMn at 1073 K and τ of 0.0039 g min ml^{-1} using a feed of 29% CH_4 in air.

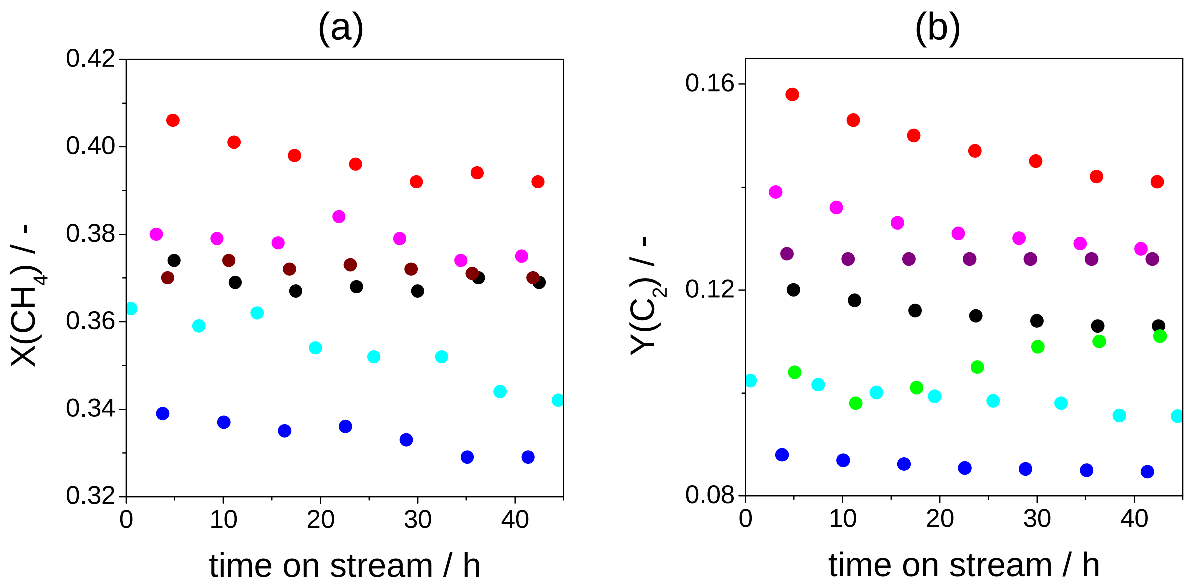

Fig. S 6 Time-on-stream (a) methane conversion and (b) yield of C_2 hydrocarbons obtained over ●-MgO, ●-MgLaLi, ●-MgLaNa, ●-MgLaCs, ●-MgLaMn, ●-MgLaBa and ●-MgLaSr at 1073 K and τ of 0.0039 g min ml^{-1} using a feed of 29% CH_4 in air.

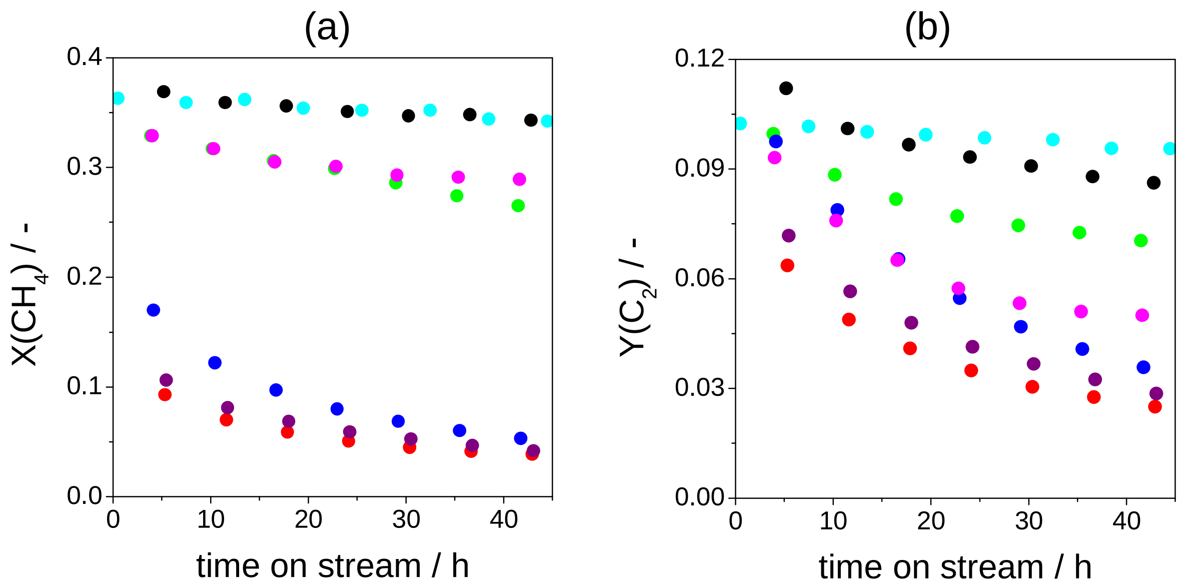

Fig. S 7 Time-on-stream (a) methane conversion and (b) yield of C_2 hydrocarbons obtained over ●-MgO, ●-MgNaLi, ●-MgNaCs, ●-MgNaMn, ●-MgLiMn, ●-MgCsLi and ●-MgCsMn at 1073 K and τ of 0.0039 g min ml^{-1} using a feed of 29% CH_4 in air.