ARTICLE

Effect of adsorbent loading on NaNiRu-DFMs' CO_2 capture and methanation: Finding optimal Na-loading using Bayesian Optimisation guided experiments

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The protocol for synthesising NiRu,XNa-DFMs(X=2.5, 5,10, and 15%)

To synthesise 5g of supported adsorbents, 2.5, 5, 10, and 15 wt% of 5 grams of the adsorbent (Na) was impregnated on 97.5, 95, 90, and 85 wt% of 5 grams of the support. The amount of Na was 0.125, 0.25, 0.5, and 0.75g, and the amount of the respective support (CeO_2-Al_2O_3) was 4.875, 4.75, 4.5, and 4.25. After impregnating the adsorbent on the support, 4g of each supported adsorbent was used to synthesise the DFMs. At this stage, 15%Ni (0.6) and 1%Ru (0.04) were impregnated on the 4g of supported adsorbents, separately. The loadings of Na for the synthesised DFMs at the end of the synthesis were 2.2, 4.3, 8.6, and 12.9 wt%.

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Electronic Supplementary Information (ESI) available: Dynamic H_2-TPSR and activity plots, GP and BO results. See DOI: 10.1039/x0xx00000x

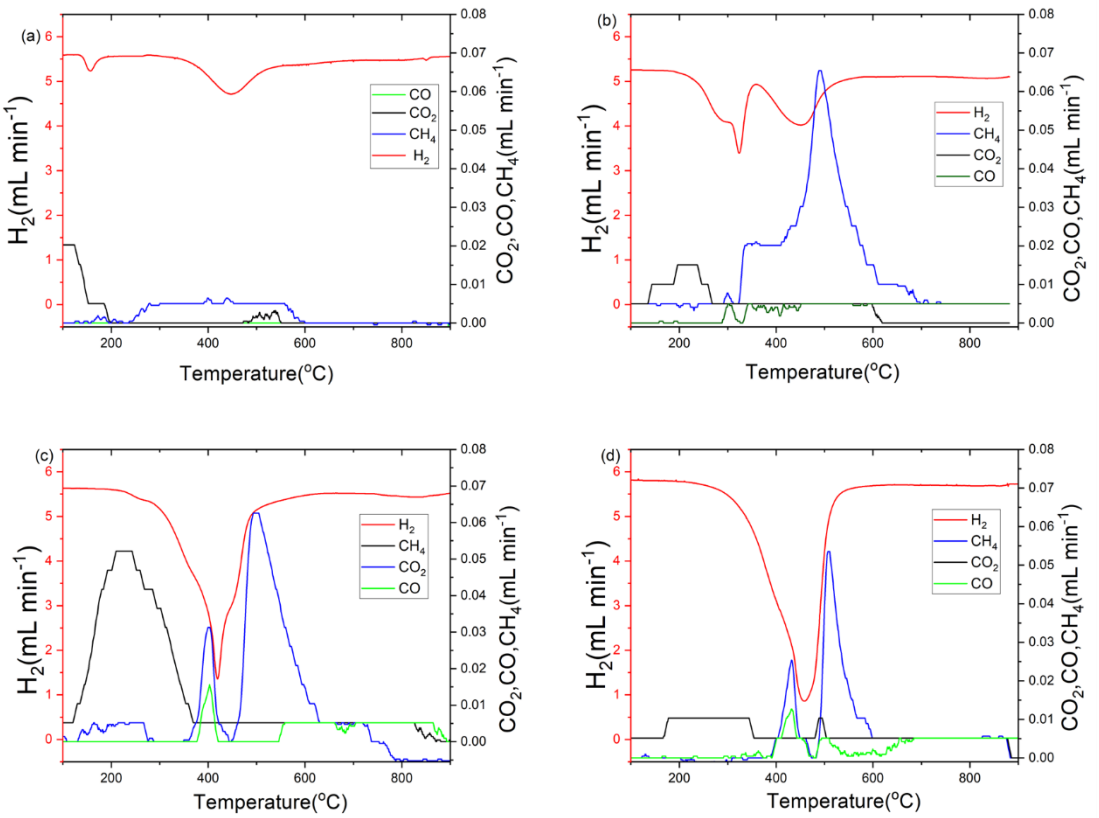
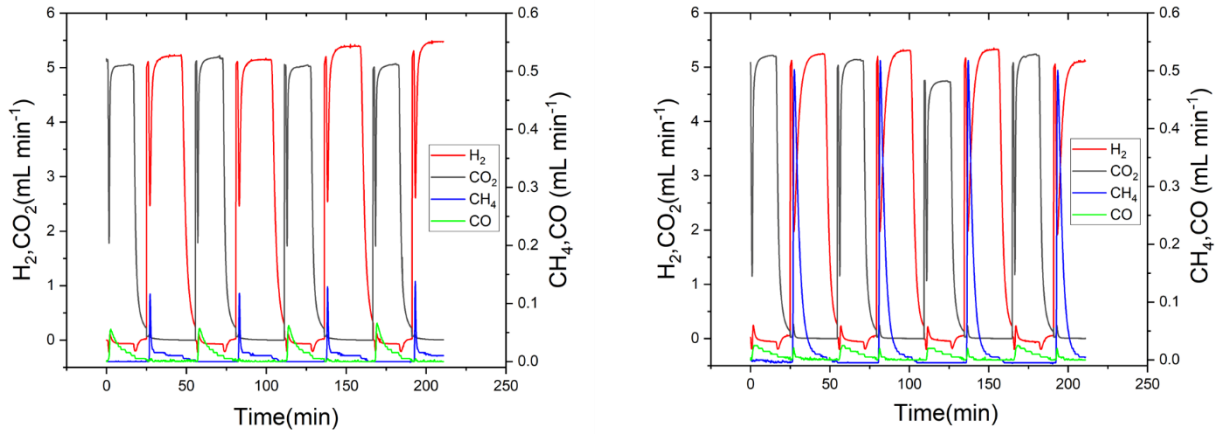


Fig. 1S dynamic curves of H2-TPSR Tests for a) 2.5Na-DFM, b) 5Na-DFM, c) 10Na-DFM, and d) 15 Na-DFM



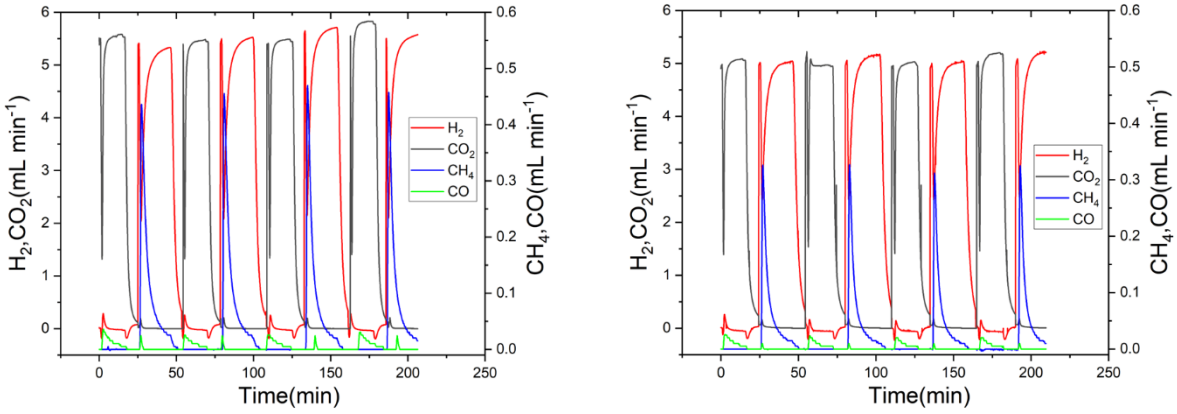


Fig. 2S dynamic curves of cyclic activity for a) 2.5Na-DFM, b) 5Na-DFM, c) 10Na-DFM, and d) 15 Na-DFM

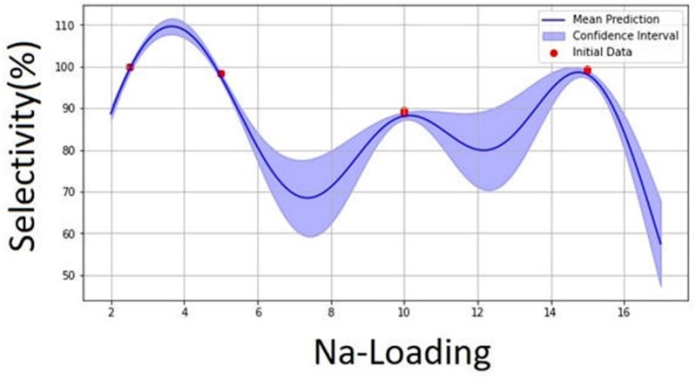


Fig. 3S selectivity towards methane predicted by BO optimised GP

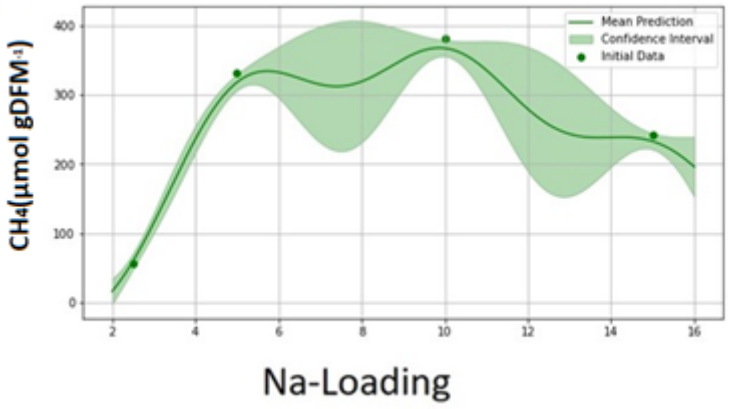


Fig. 4S produced methane predicted by BO optimised GP

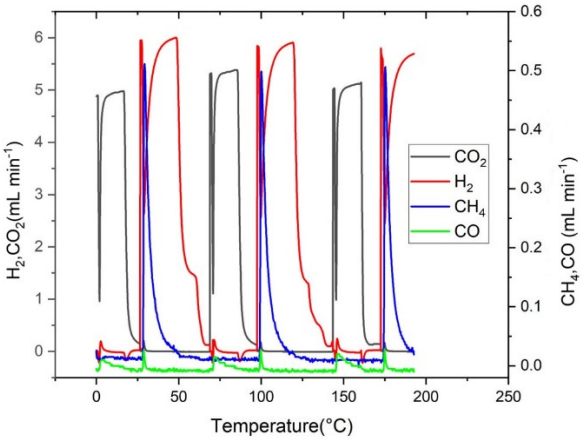


Fig. 5S dynamic curves of cyclic activity of 7.9Na-DFM

Reference XRD Patterns

Al_2O_3, Reference Code: 00-001-1303

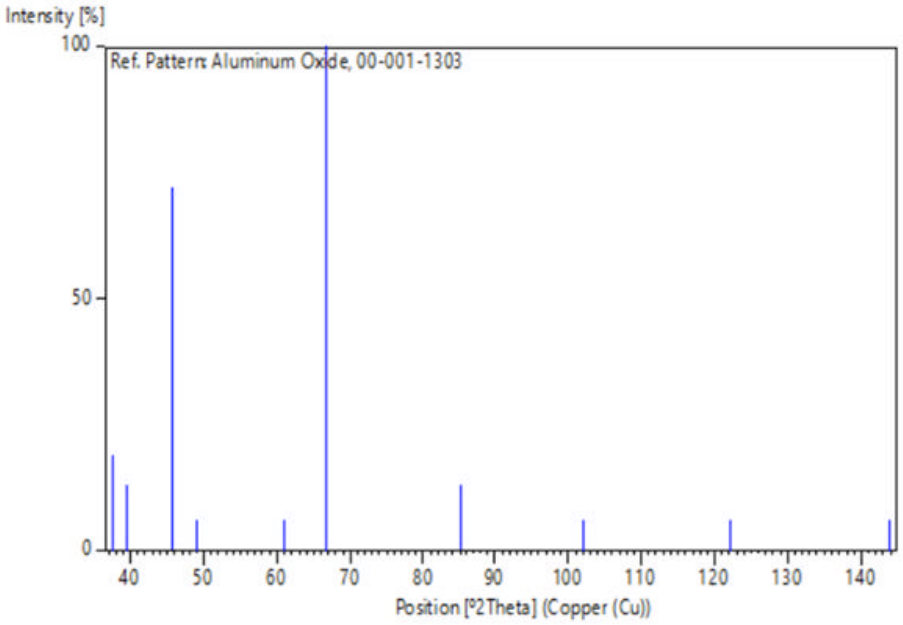


Fig. 6S reference XRD pattern for Al_2O_3

CeO_2, Reference Code: 03-065-5923

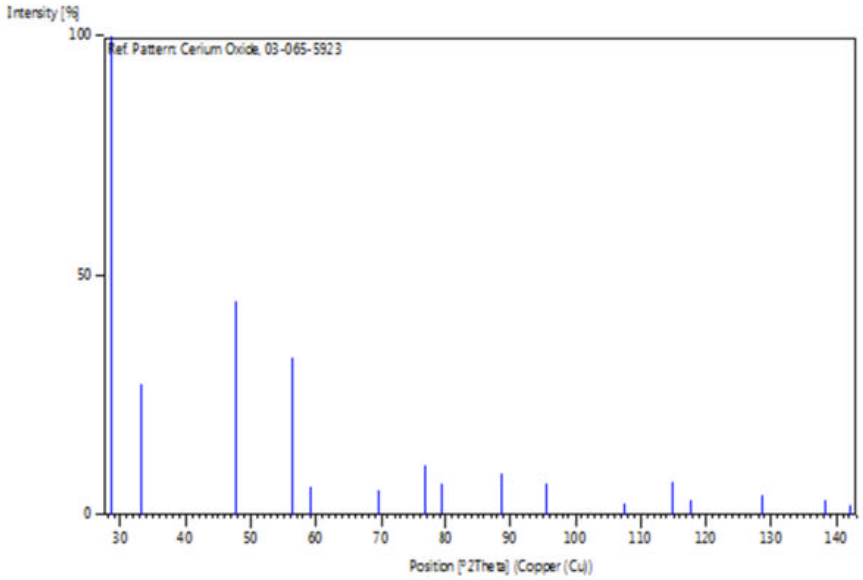


Fig. 7S reference XRD pattern for CeO_2

NiO, Reference Code: 00-047-1049

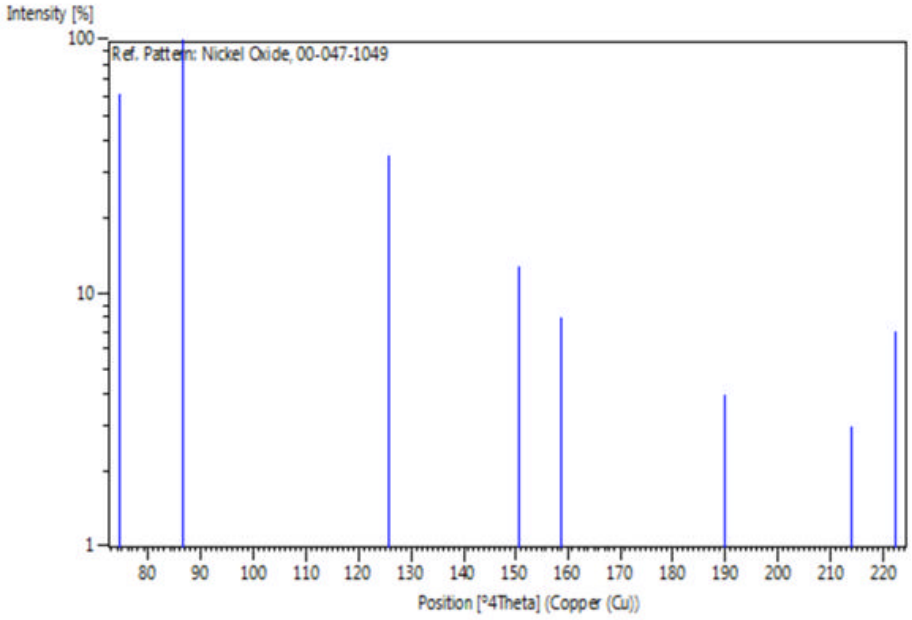


Fig. 8S reference XRD pattern for NiO

$NaNO_3$

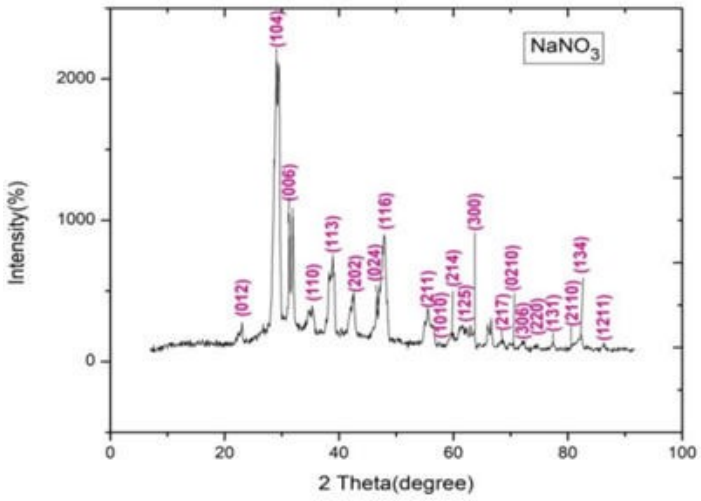


Fig. 9S reference XRD pattern for $NaNO_3$, Open access[1]

Mathematical Background of GP and BO

A Gaussian Process (GP) is a collection of random variables, any finite subset of which follows a multivariate Gaussian distribution. A GP is specified by its mean function and the covariance function (Kernel). In this study, we considered the mean function as zero and the Radial Based Function (RBF) kernel was utilised in GP modelling.[2]

$f(x) \sim GP(m(x), k(x, x'))$

Where $m(x)$, =0

$k(x, x')$= Kernel

RBF kernel: $k(x, x') = \sigma^2 \exp(-\frac{\|x-x'\|^2}{2l^2})$

where:

- σ^2 is the signal variance
- l is the length scale

The acquisition function in Bayesian optimisation determines the next recommended point. Herein we used Expected Improvement (EI) as the acquisition function.[3]

$EI(x) = E[\max(0, f(x)) - f(x^+)]$

Where $f(x^+)$ is the best observed value so far.

The next point to evaluate is:

$x_{next} = arg \max_x a(x)$

Safety and Hazards

The synthesis of the materials requires the usage of hazardous materials. All the materials safety sheets must be read before usage, and proper PPE should be used while preparing the DFMs. Testing DFMs requires intermediate or high temperatures, and it can produce toxic gases (CO, NOx). All safety issues should be considered before starting the experiments, such as leak testing, directing gases to the exhaust, and cooling down the setup after the experiments. Appropriate PPE must be worn during all experiments. For some characterisation tests like XRD, special safety precautions must be considered due to the usage of the X-ray beam.

References

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