**Dataset for:** **The reactivity of lattice nitrogen within the Ni2Mo3N and NiCoMo3N phases**

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Fig 2 data (Microsoft Excel format) contains XRD patterns before and after catalytic reactions for Ni2Mo3N and NiCoMo3N.

Fig 5 data (Microsoft Excel format) contains nitrogen isotopomer distributions calculated from heterolytic temperature programmed nitrogen isotopic exchange (TPNIE) on Ni2Mo3N and NiCoMo3N.

The remaining data are neutron diffraction collected with the ILL D20 diffractometer under conditions as shown in the table.

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| --- | --- | --- | --- | --- | --- | --- |
| **Run number and material** | **Gas** | **T / °C** | ***a* / Å** | **N site occupancy** | **Full details in SI** | **Data set number** |
| 1. Ni2Mo3N | H2/N2 | Ambient | 6.63050(9) | 0.973(7) | Fig. S2a | 930038 |
|  | H2/N2 | 700 | 6.66794(10) | 0.975(5) | Fig. S2b | 930071 |
|  | H2/N2 | 400 | 6.65045(10) | 0.974(7) | Fig S2c | 930089 |
| 2. NiCoMo3N | H2/N2 | Ambient | 6.63680(9) | 0.966(6) | Fig. S3a | 930203 |
|  | H2/N2 | 700 | 6.67252(10) | 0.949(5) | Fig. S3b | 930229 |
|  | H2/N2 | 400 | 6.65547(10) | 0.959(5) | Fig. S3c | 930253 |
| 3. Ni2Mo3N | H2/N2 | 700 | 6.66484(13) | 0.960(8) | Fig. S4a | 930115 |
|  | H2/Ar | 700 | 6.66454(13) | 0.957(8) | Fig. S4b | 930125 |
|  | H2/N2 | 700 | 6.66494(13) | 0.960(8) | Fig. S4c | 930139 |
| 4. NiCoMo3N | H2/N2 | Ambient | 6.63655(9) | 0.967(5) | Fig. S5a | 930330 |
|  | H2/N2 | 700 | 6.67326(11) | 0.953(5) | Fig. S5b | 930345 |
|  | H2/Ar | 700 | 6.67268(11) | 0.947(5) | Fig. S5c | 930357 |
|  | H2/N2 | 700 | 6.67332(10) | 0.956(5) | Fig. S5d | 930369 |
| 5. Ni2Mo3N | H2/N2 | Ambient | 6.62841(10) | 0.983(7) | Fig. S6a | 930140 |
|  | H2/N2 | 700 | 6.66509(11) | 0.965(7) | Fig. S6b | 930166 |
|  | H2/Ar | 700 | 6.66465(11) | 0.956(7) | Fig. S6c | 930178 |
|  | H2/Ar | 400 | 6.64725(10) | 0.966(7) | Fig. S6d | 930190 |
|  | H2/N2 | 400 | 6.64761(10) | 0.968(7) | Fig. S6e | 930202 |
| 6. NiCoMo3N | H2/N2 | Ambient | 6.63574(10) | 0.963(6) | Fig. S7a | 930257 |
|  | H2/N2 | 700 | 6.67554(12) | 0.948(6) | Fig. S7b | 930283 |
|  | H2/Ar | 700 | 6.67496(11) | 0.945(5) | Fig. S7c | 930295 |
|  | H2/Ar | 400 | 6.65567(10) | 0.955(5) | Fig. S7d | 930307 |
|  | H2/N2 | 400 | 6.65634(10) | 0.957(5) | Fig. S7e | 930319 |

Data files are the data set number given in GSAS format with a .gsa extension

GSAS parameter file for D20 also supplied: D20.prm