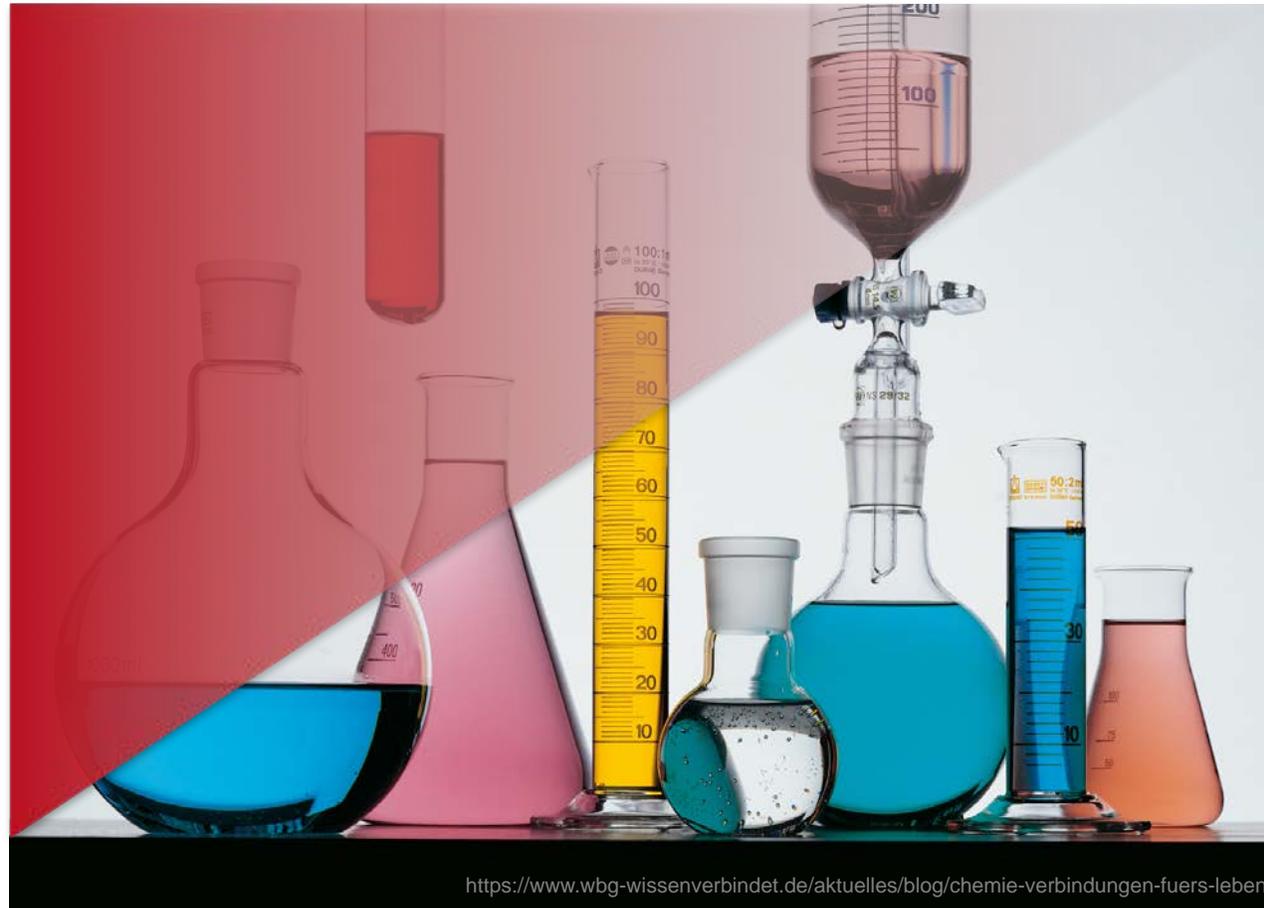


Theoretische Chemie: Von der Physik zur Katalyse

Prof. Dr. Vera Krewald
vera.krewald@tu-darmstadt.de

Einweihung
des Lichtenberg II Hochleistungsrechners
11.07.2023

Chemie



Chemie

“knallt und stinkt”

“nicht natürlich”

“gefährlich”

“giftig”

“Umweltverschmutzung”

“forever chemicals”



Chemie

“knallt und stinkt”

“nicht natürlich”

“gefährlich”

“giftig”

“Umweltverschmutzung”

“forever chemicals”



Chemie kann duften

Chemie im Leben

Chemie verstehen

... für die Medizin

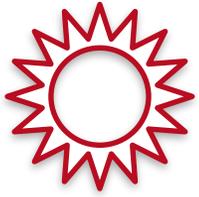
... für die Umwelt

... für die Zukunft

Chemie

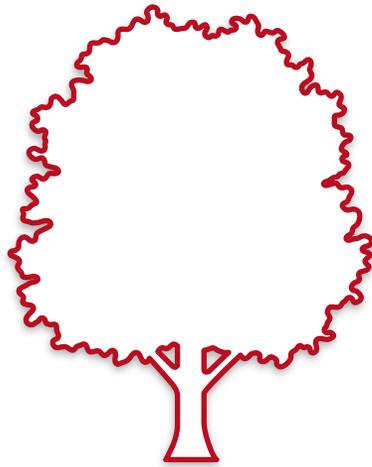
Stoffe verstehen und umwandeln

Chemie



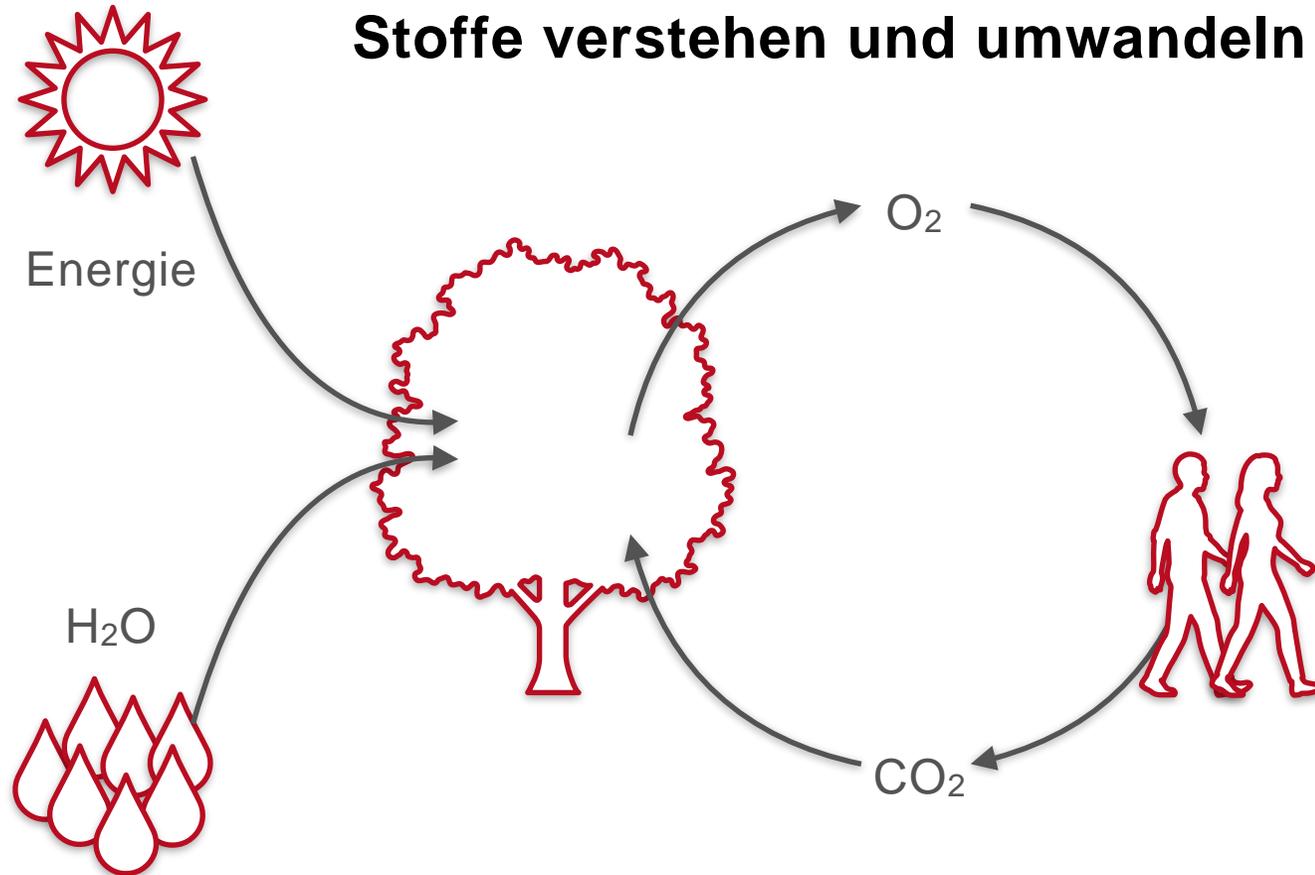
Energie

Stoffe verstehen und umwandeln



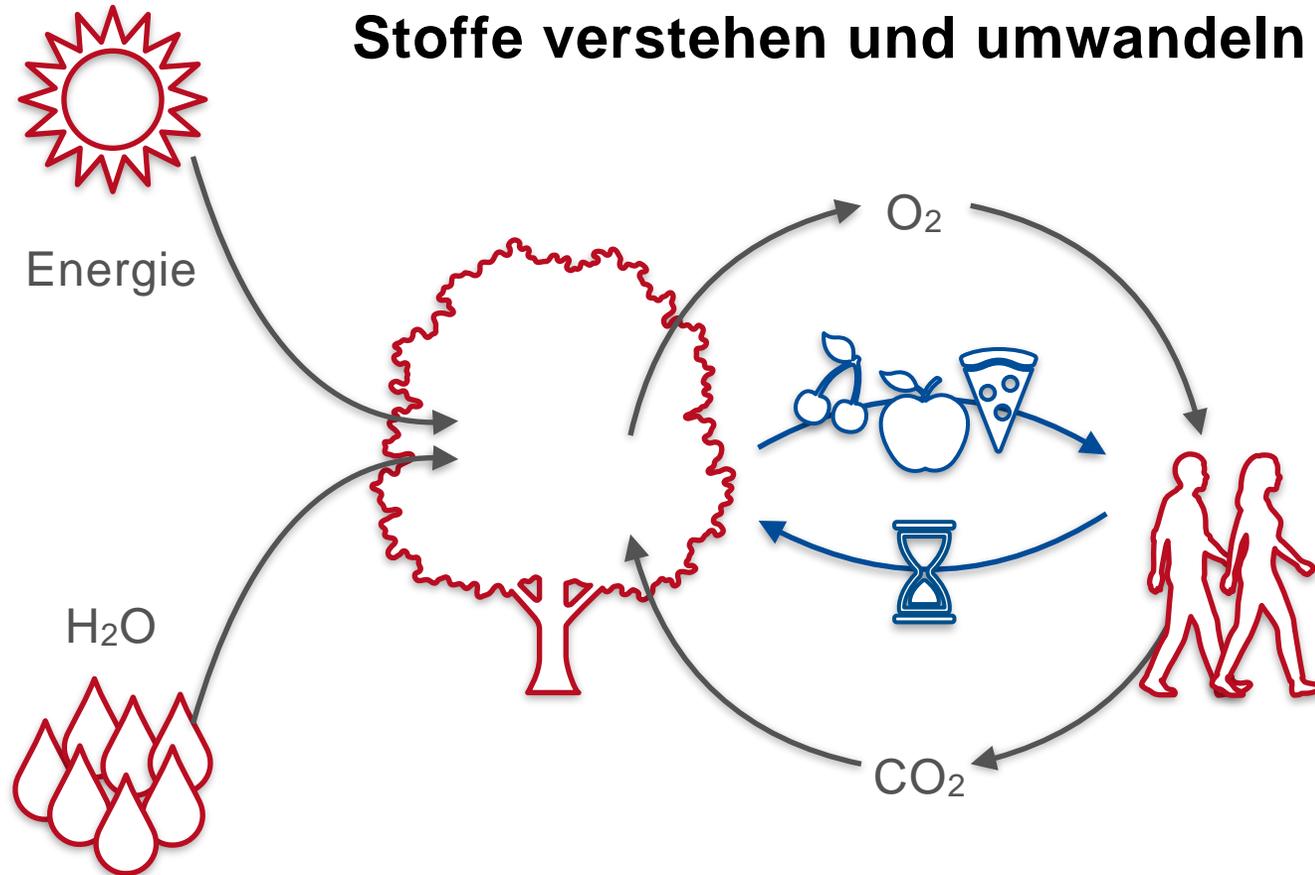
Chemie

Stoffe verstehen und umwandeln



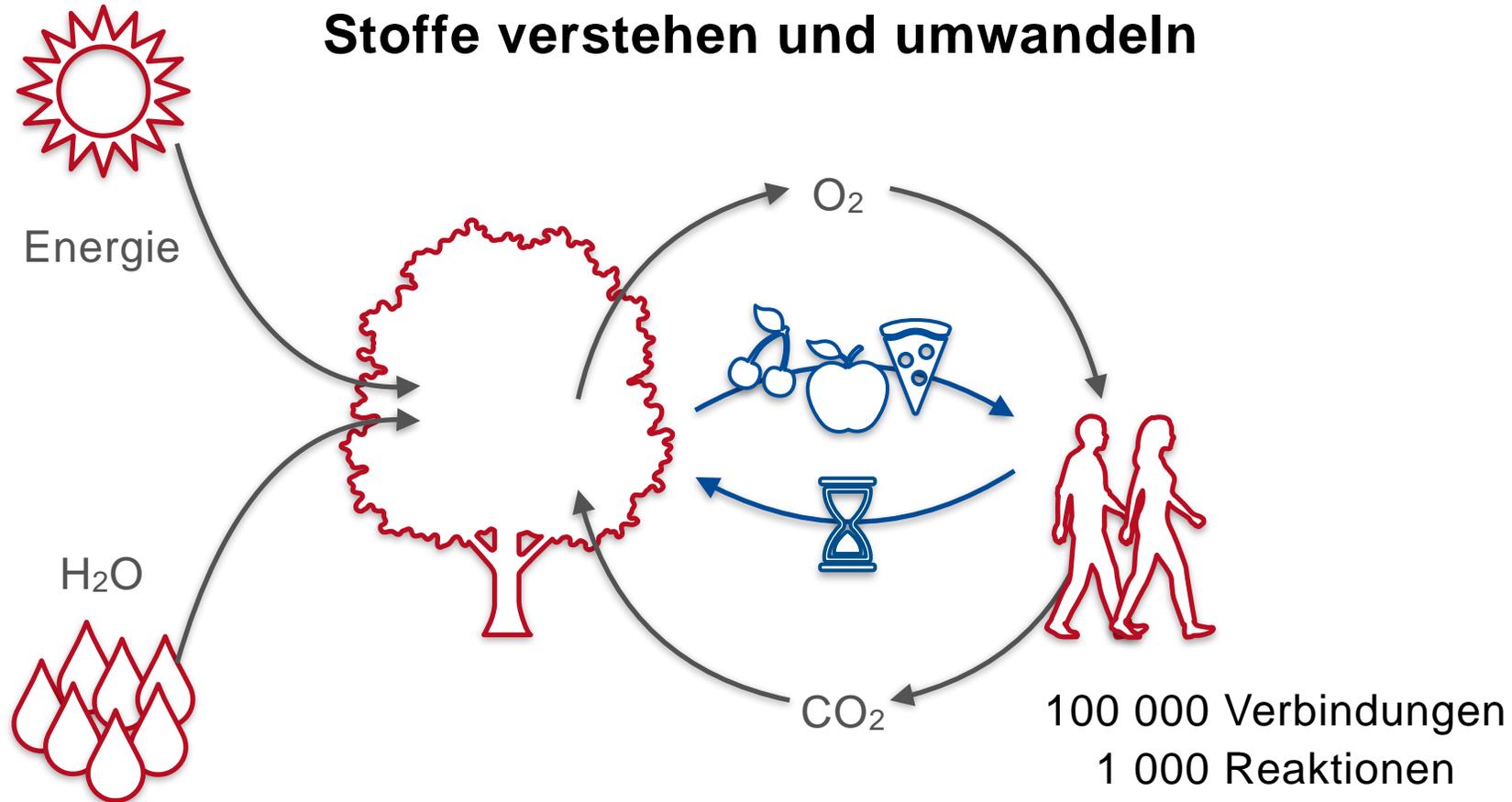
Chemie

Stoffe verstehen und umwandeln



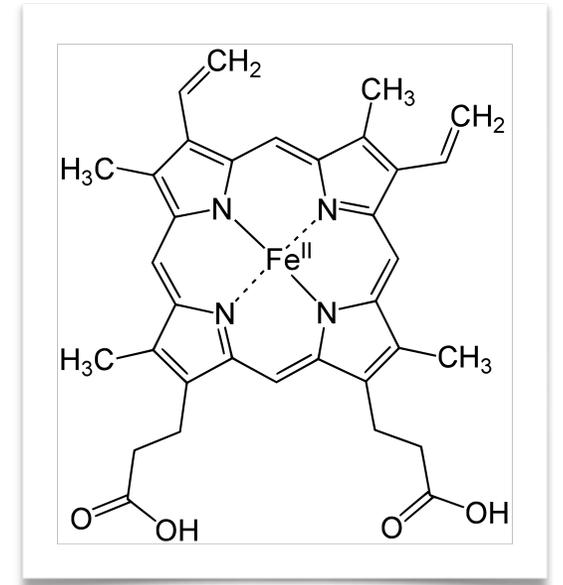
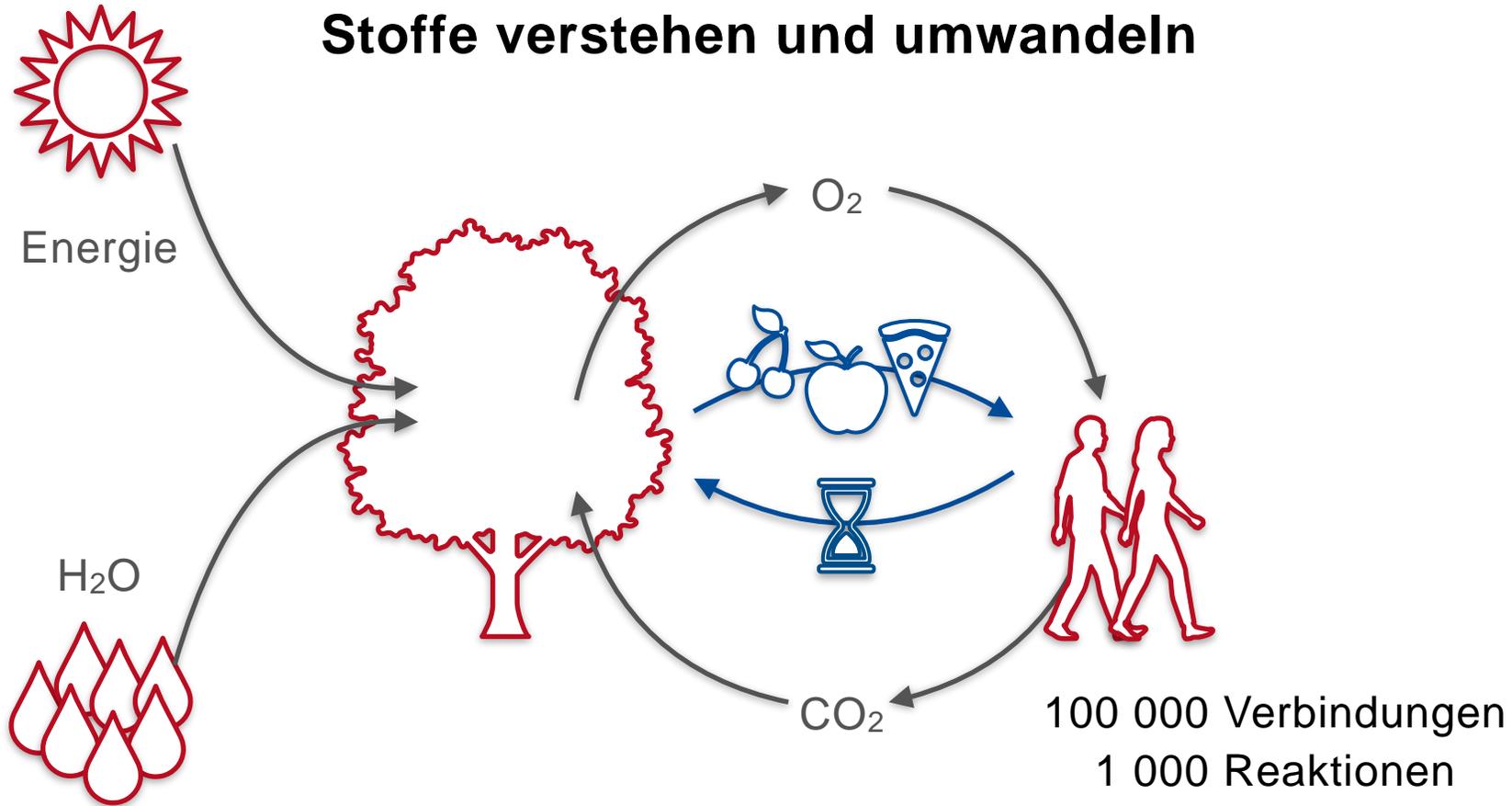
Chemie

Stoffe verstehen und umwandeln



Chemie

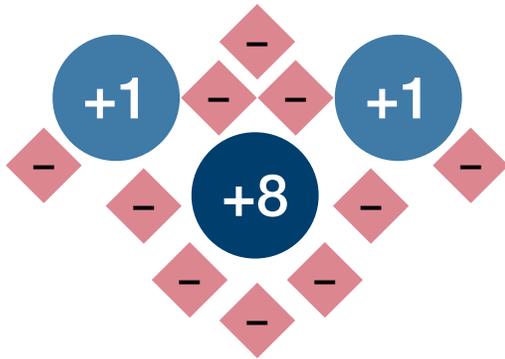
Stoffe verstehen und umwandeln



Häm b im Hämoglobin

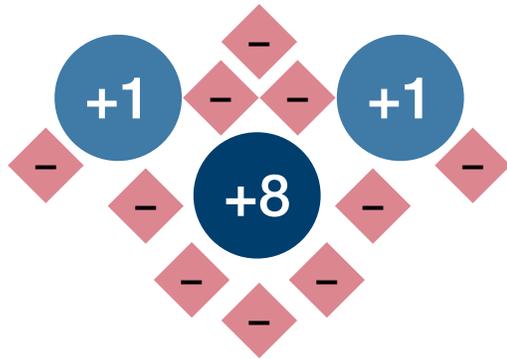
Theoretische Chemie

Physik
Gesetze

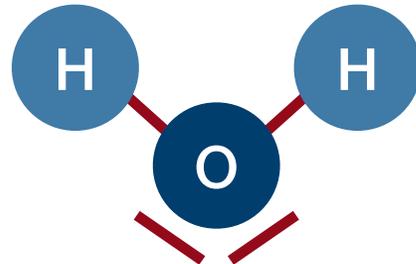


Theoretische Chemie

Physik
Gesetze



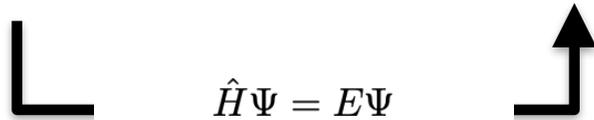
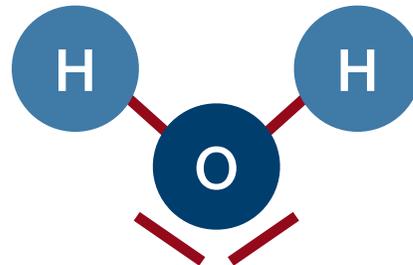
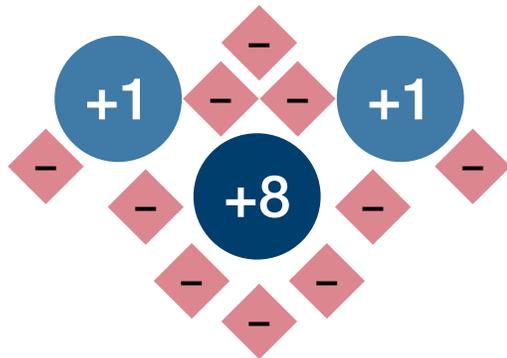
Chemie
Konzepte



Theoretische Chemie

Physik
Gesetze

Chemie
Konzepte



$$f(1) = h(1) + \sum_b J_b(1) - K_b(1)$$

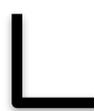
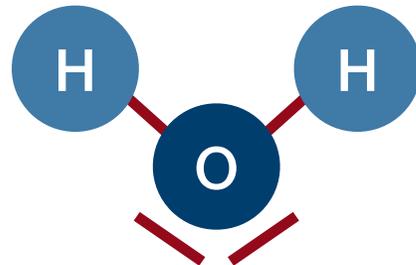
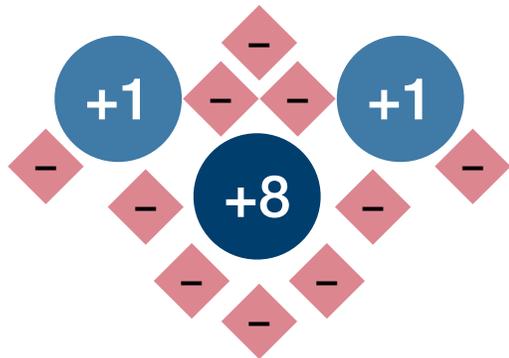
$$E = \sum_i^{\text{occ}} \langle i|h|i \rangle + 1/2 \sum_i^{\text{occ}} \sum_j^{\text{occ}} \langle ij||ij \rangle$$



Theoretische Chemie

Physik
Gesetze

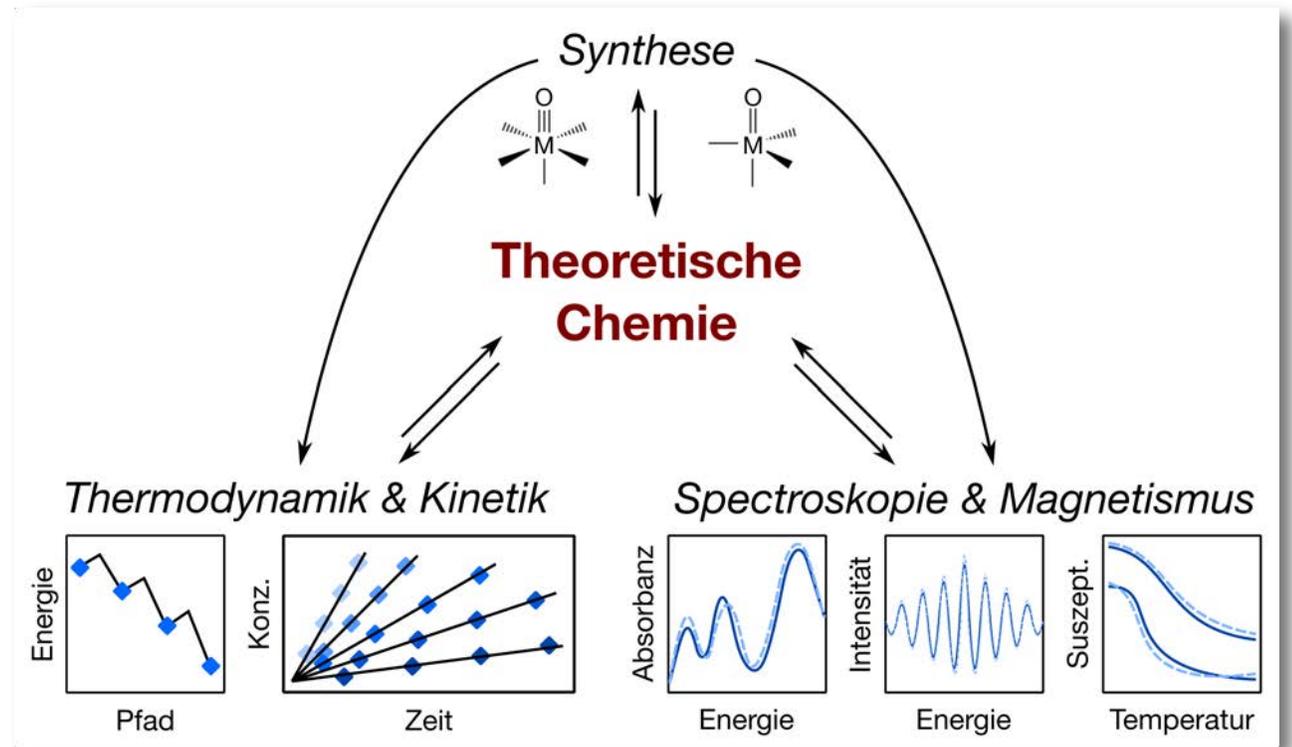
Chemie
Konzepte



$$\hat{H}\Psi = E\Psi$$

$$f(1) = h(1) + \sum_b J_b(1) - K_b(1)$$

$$E = \sum_i^{\text{occ}} \langle i|h|i \rangle + 1/2 \sum_i^{\text{occ}} \sum_j^{\text{occ}} \langle ij||ij \rangle$$



Theoretische Chemie



Schrödinger Slater Hartree Roothaan



Born

**Pauli & Bohr
und viele mehr**

Kohn

Theoretische Chemie



Schrödinger Slater Hartree Roothaan



Born

**Pauli & Bohr
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Kohn

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$$f(1) = h(1) + \sum_b J_b(1) - K_b(1)$$

$$E = \sum_i^{\text{occ}} \langle i|h|i \rangle + 1/2 \sum_i^{\text{occ}} \sum_j^{\text{occ}} \langle ij||ij \rangle$$

```
CALL QENTER('FMAT')

NDIM2M=(NS...
IDISK=0
CALL IDAFI...

IFSTA=1
DO ISYR=1,1
  NBR=NORB
  IF(NBR.E...
  NB3=(NBR...
  NBNB=NBR...
  ISYS=ISY...
  IS3RS=(I...
  DO ISYP=...
  NIP=NI...
  NOP=NO...
  NAESP=...
  IF(NOP...
  ISYQ=I...
  IS3PQ=...

import cmd
import pymol
from cmd import _cmd,lock,unlock,Shortcut
_feedback,fb_module,fb_mask,is_list
DEFAULT_ERROR, DEFAULT_SUCCESS, _ra

def resume(fname):
  r = DEFAULT_ERROR
  if os.path.exists(fname):
    if(re.search(r"\.py$|\.PYS$|\.pym$"):
      r = cmd.do("run %s"%fname)
    else:
      r = cmd.do("@%s"%fname)
  if is_ok(r):
    r = cmd.do("log_open %s,a"%fname)
  if _raising(r): raise pymol.CmdExcept
  return r

def log_open(fname='log.pml',mode='w'):
  try:
    try:
      if hasattr(pymol,"_log_file")
      if pymol._log_file!=None:
        pymol._log_file.close
        del pymol._log_file
    except:
      pass
```



Theoretische Chemie

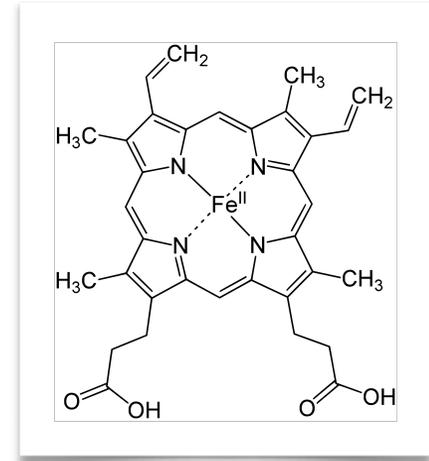


Schrödinger Slater Hartree Roothaan

$$\hat{H}\Psi = E\Psi$$

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```
CALL QENTER('FMAT')

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IDISK=0
import pymol
CALL IDAFI...
from cmd import _cmd,lock,unlock,Shortcut
_feedback,fb_module,fb_mask,is_list
DEFAULT_ERROR, DEFAULT_SUCCESS, _ra

IFSTA=1
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NBNB=NBR
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DO ISYP=
NIP=NI
NOP=NO
NAESP=
IF(NOP
ISYQ=I
IS3PQ=

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        else:
            r = cmd.do("@%s"%fname)
    if is_ok(r):
        r = cmd.do("log_open %s,a"%fname)
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        try:
            if hasattr(pymol,"_log_file")
                if pymol._log_file!=None:
                    pymol._log_file.close
                    del pymol._log_file
        except:
            pass
```



Born Pauli & Bohr Kohn

und viele mehr



Theoretische Chemie

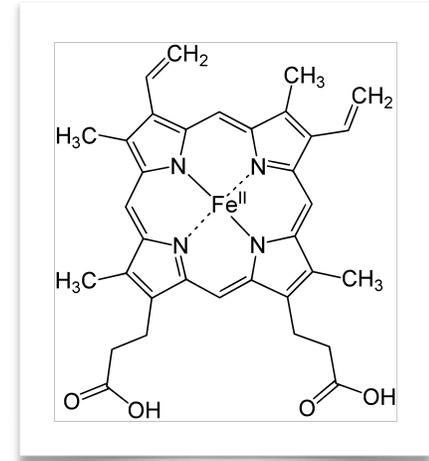


Schrödinger Slater Hartree Roothaan

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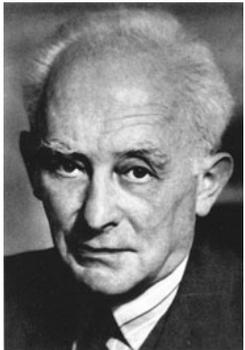
```
CALL CENTER('FMAT')

NDIM2M=(NS...
import cmd
IDISK=0
import pymol
CALL IDAFI...
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_feedback,fb_module,fb_m...
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        if _raising(r): raise pymo
    return r

def log_open(fname='log.pml',m
try:
    try:
        if hasattr(pymol,"
            if pymol._log_
                pymol._log_
                del pymol.
    except:
        pass
```



Born



**Pauli & Bohr
und viele mehr**



Kohn



Stoffe verstehen

Eigenschaften vorhersagen:

Physikalische Gesetze in
chemische Konzepte
übersetzen

Stoffe verstehen

Eigenschaften vorhersagen:

Physikalische Gesetze in
chemische Konzepte
übersetzen

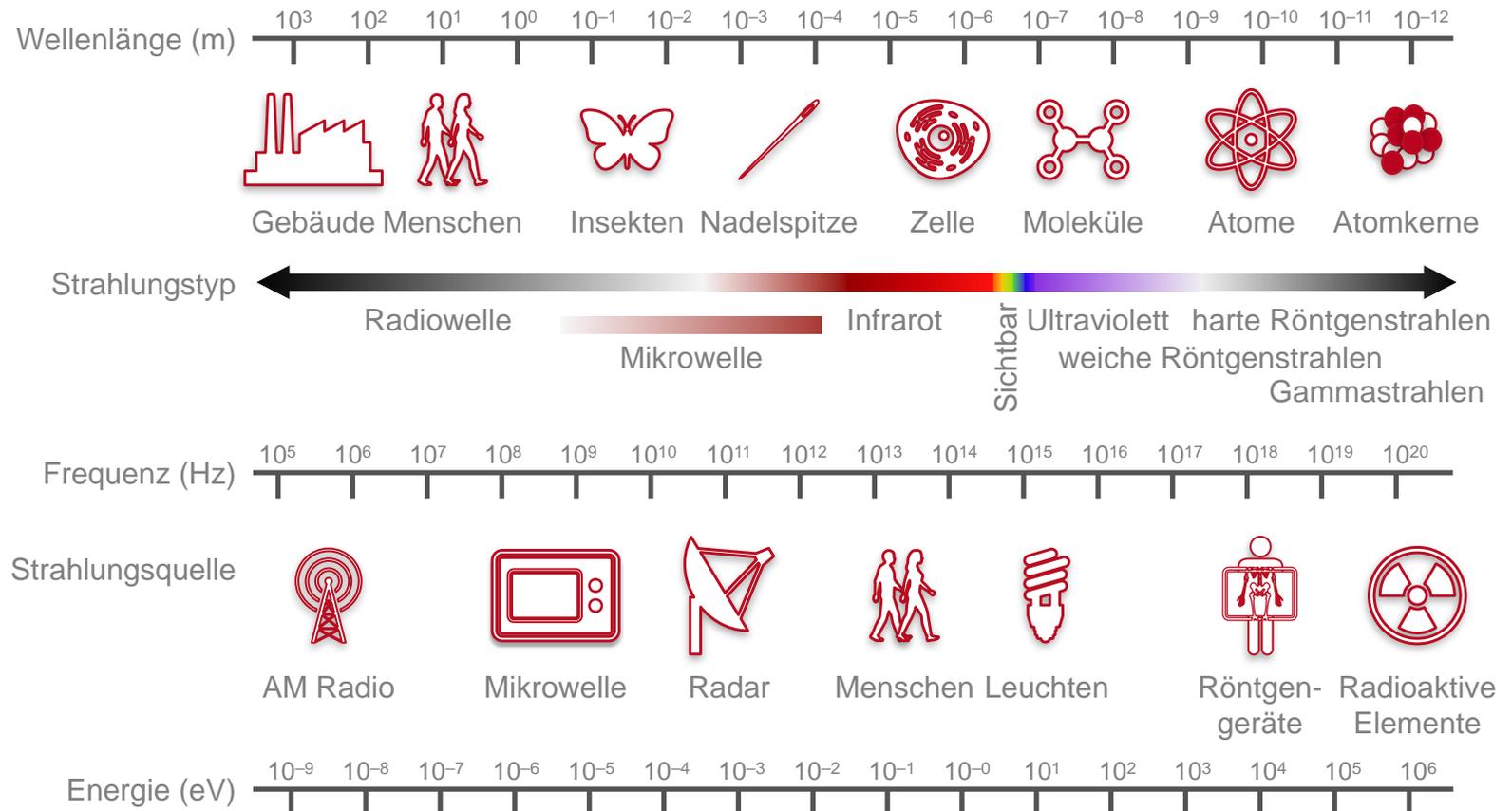
– Struktur eines Moleküls

Stoffe verstehen

Eigenschaften vorhersagen:

Physikalische Gesetze in
chemische Konzepte
übersetzen

- Struktur eines Moleküls
- Optische, vibronische,
magnetische Eigenschaften



Stoffe verstehen und umwandeln



Eigenschaften vorhersagen:

Physikalische Gesetze in
chemische Konzepte
übersetzen

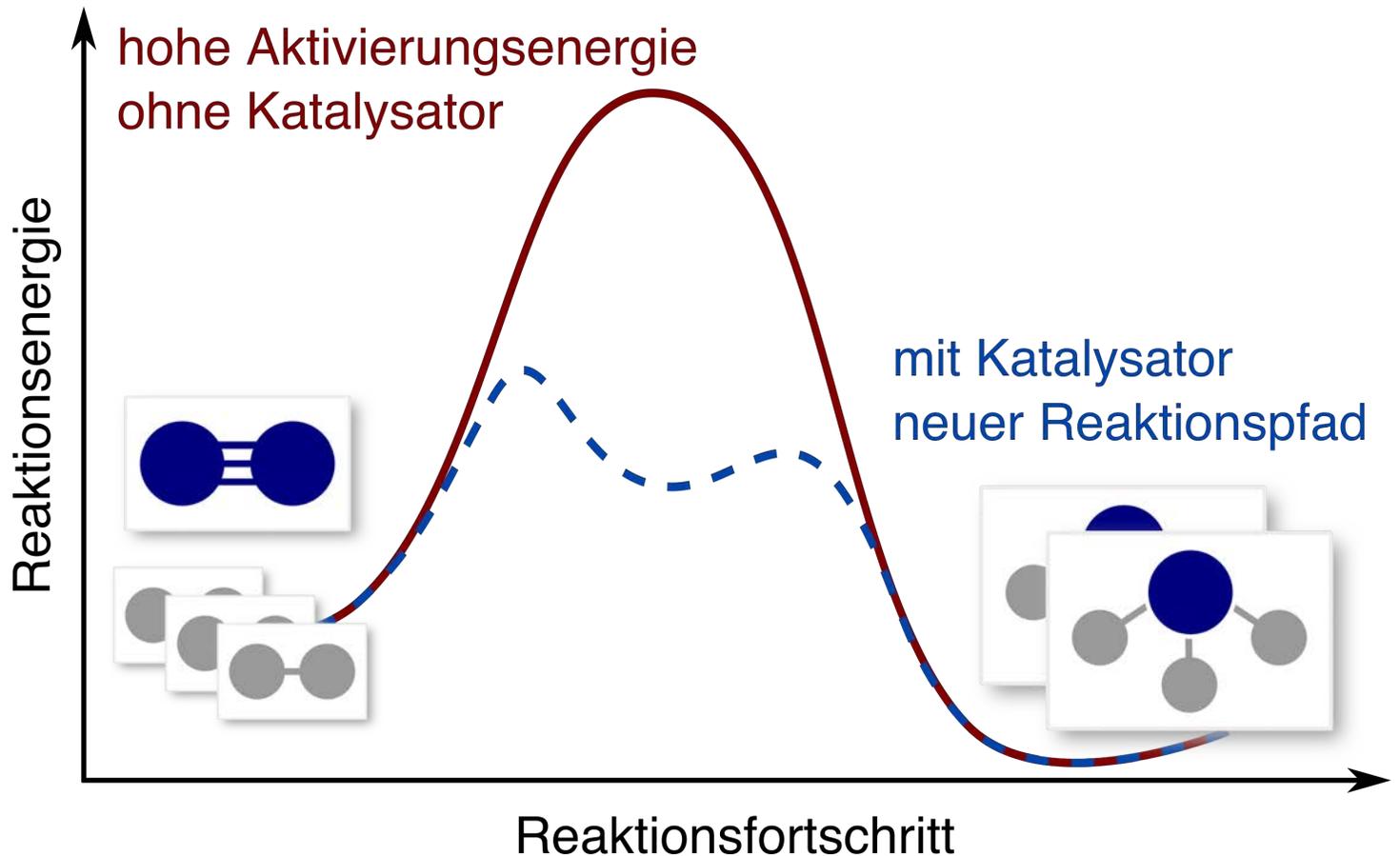
- Struktur eines Moleküls
- Optische, vibronische,
magnetische Eigenschaften

Stoffe verstehen und umwandeln

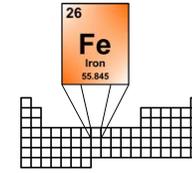
Eigenschaften vorhersagen:

Physikalische Gesetze in
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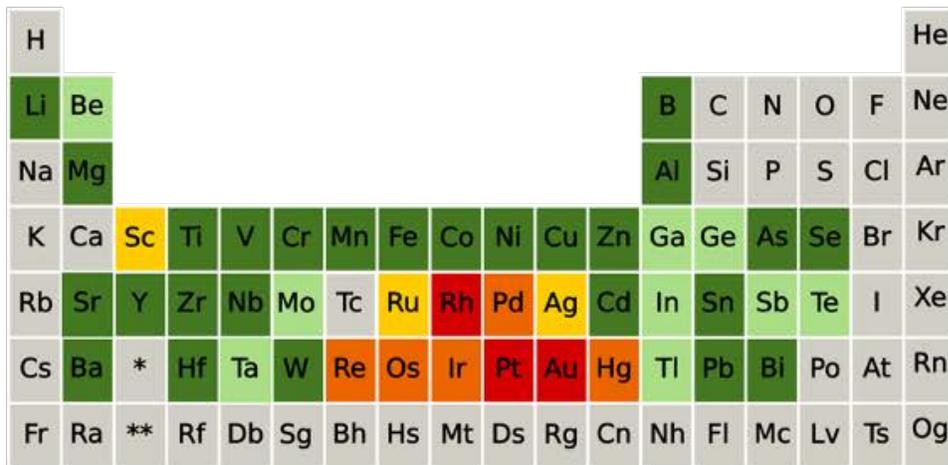
- Struktur eines Moleküls
- Optische, vibronische,
magnetische Eigenschaften
- Reaktivität und **Katalyse**



Chemie und Katalyse für eine grünere Zukunft

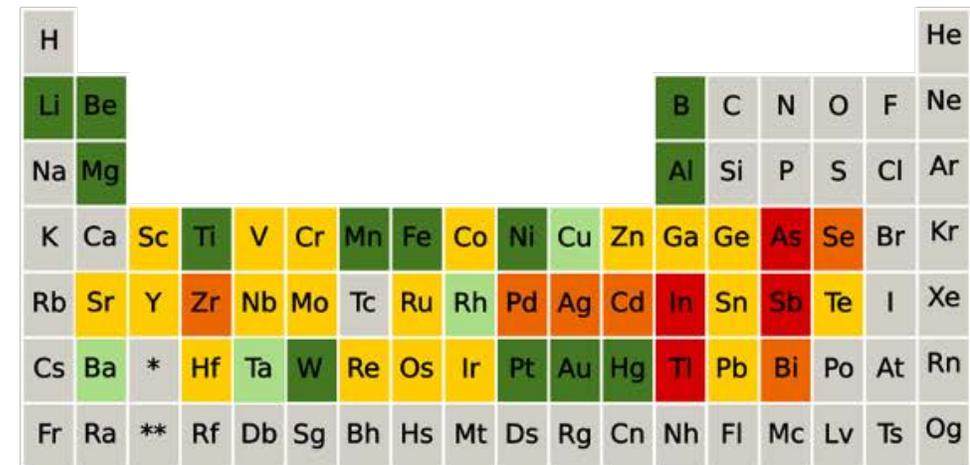


Umwelteinfluss



*	La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
**	Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

Liefersicherheit

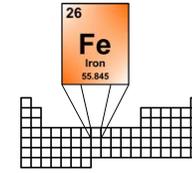


*	La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
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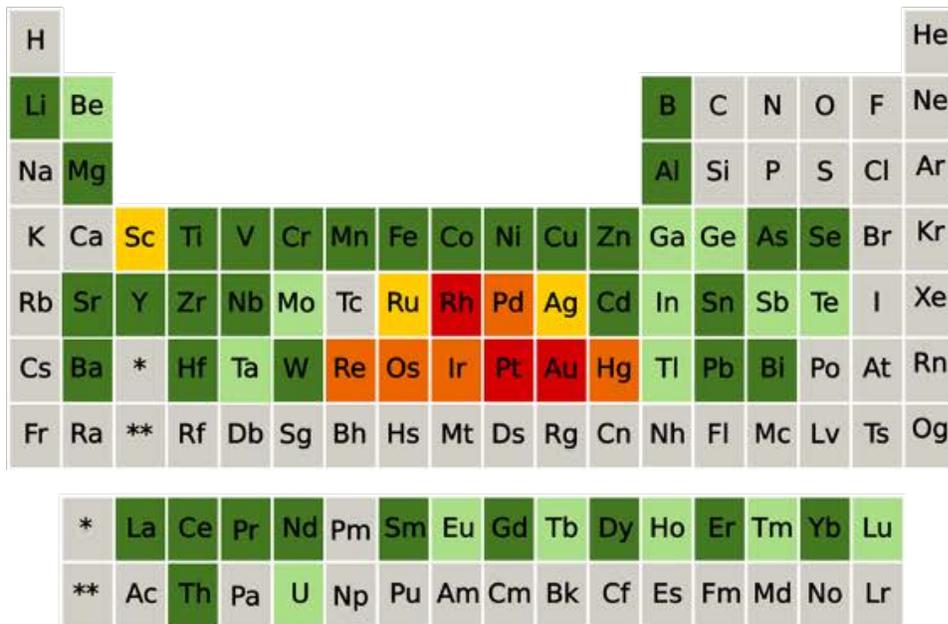


Adapted by U. I. Kramm from Graedel *et al.*, „Criticality of metals and metalloids“, *PNAS* 2015, 112, 4257.

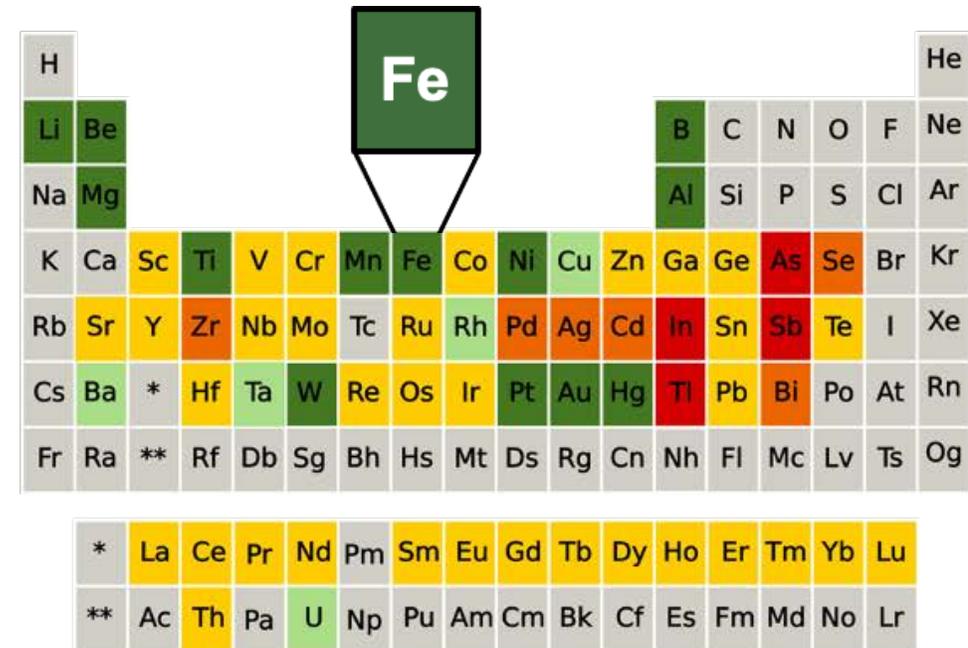
Chemie und Katalyse für eine grünere Zukunft



Umwelteinfluss

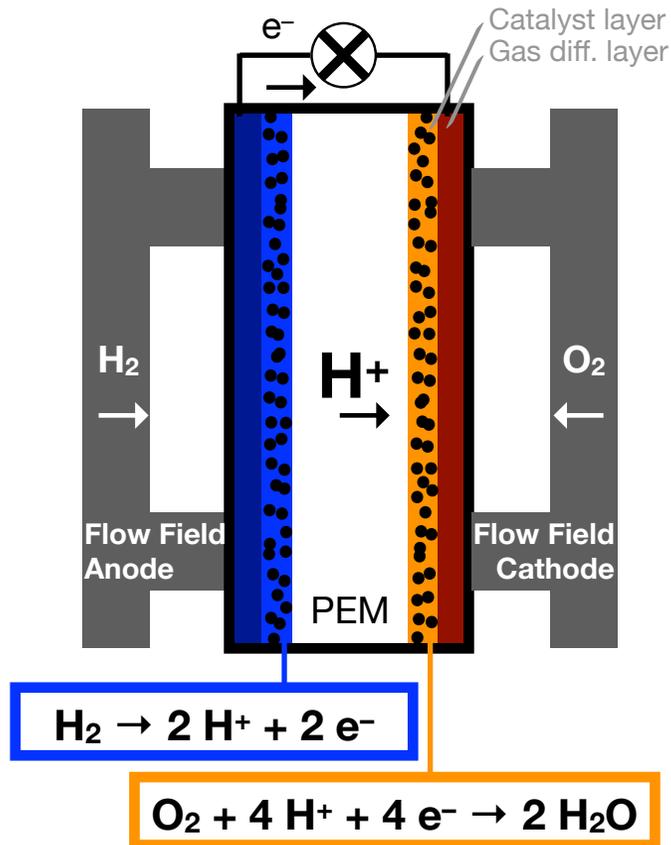
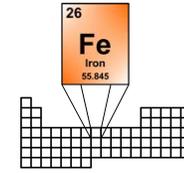


Liefersicherheit



Adapted by U. I. Kramm from Graedel *et al.*, „Criticality of metals and metalloids“, *PNAS* 2015, 112, 4257.

Katalyse mit Eisen für eine grünere Zukunft



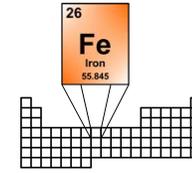
Motivation

- Sauerstoffreduktion: Relevanz für grüne Mobilität
- Platin: effizient, stabil, teuer, kritisch
- Eisen: effizient, weniger stabil, günstiger, grün

Fragestellungen

- Identifikation des aktiven Zentrums
- Verständnis des Reaktionsmechanismus
- Verbesserung der Katalysatorstabilität

Katalyse mit Eisen für eine grünere Zukunft



Motivation

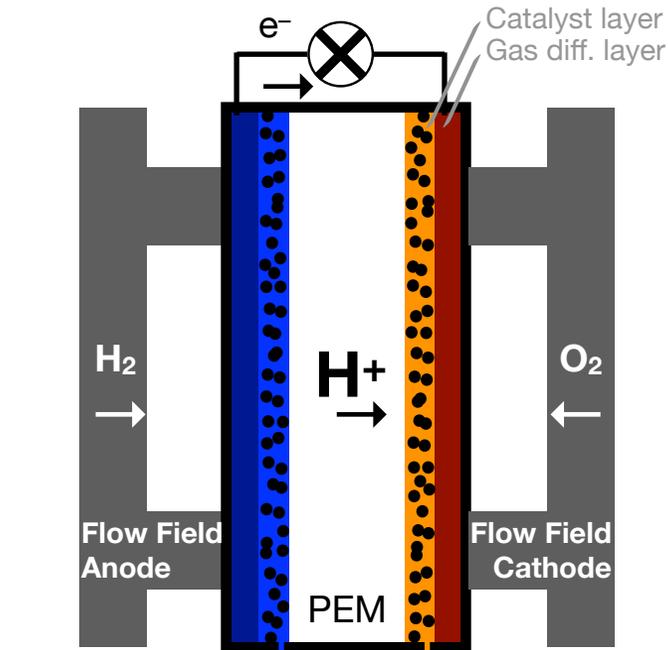
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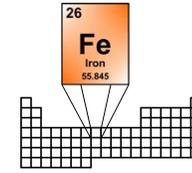
Katalysatormaterial

amorph, heterogen



In collaboration with Prof. U. I. Kramm

Katalyse mit Eisen für eine grünere Zukunft



Motivation

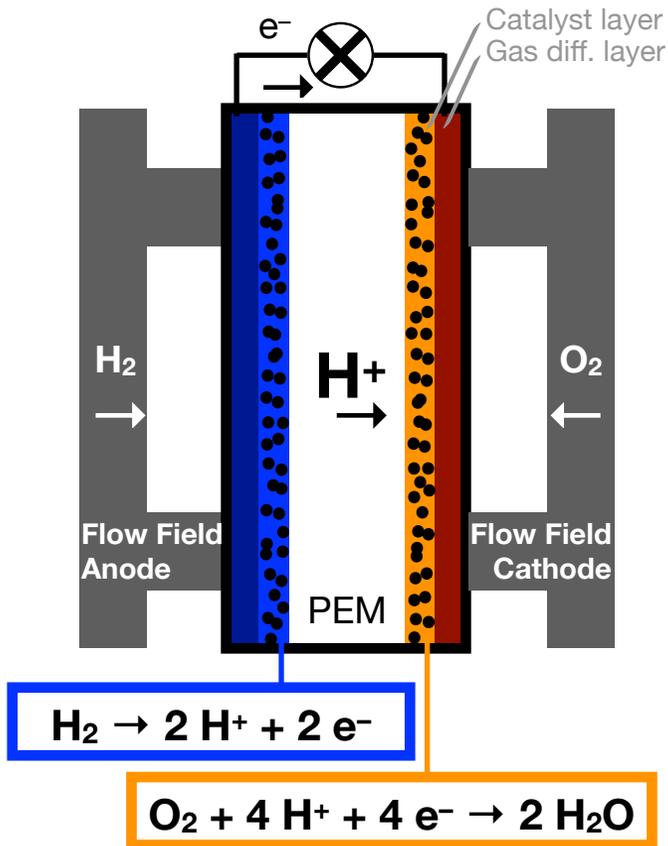
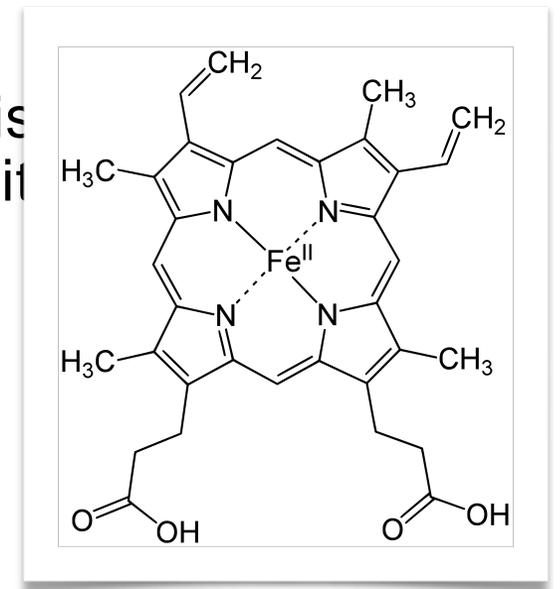
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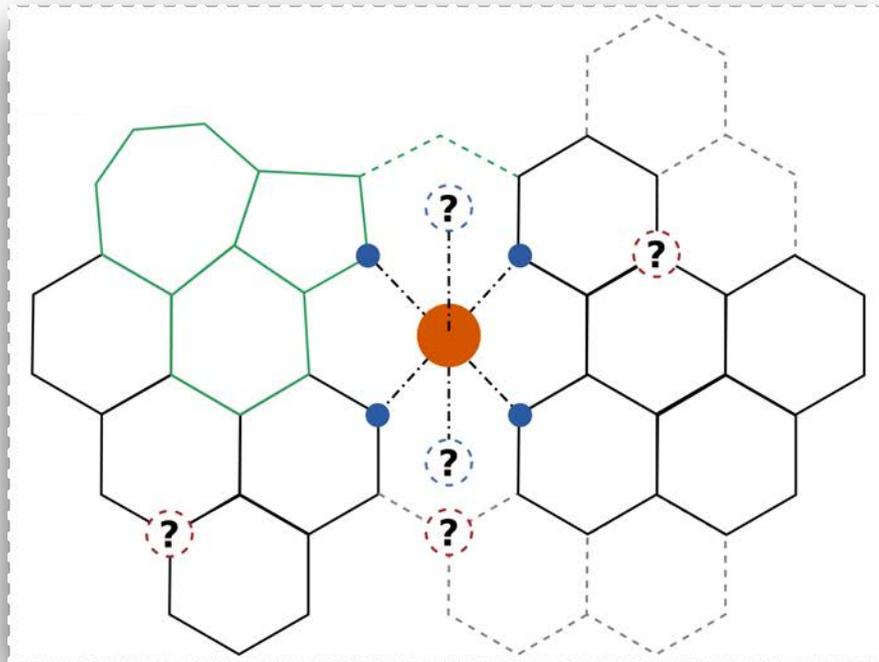
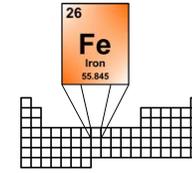
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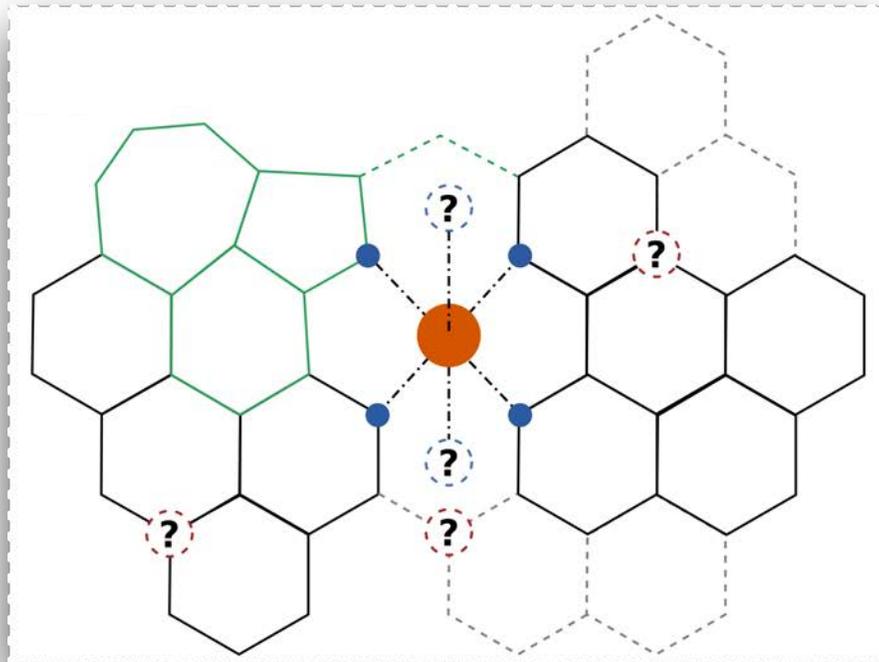
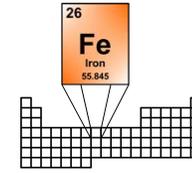
In collaboration with Prof. U. I. Kramm

Das katalytisch aktive Eisen-Zentrum



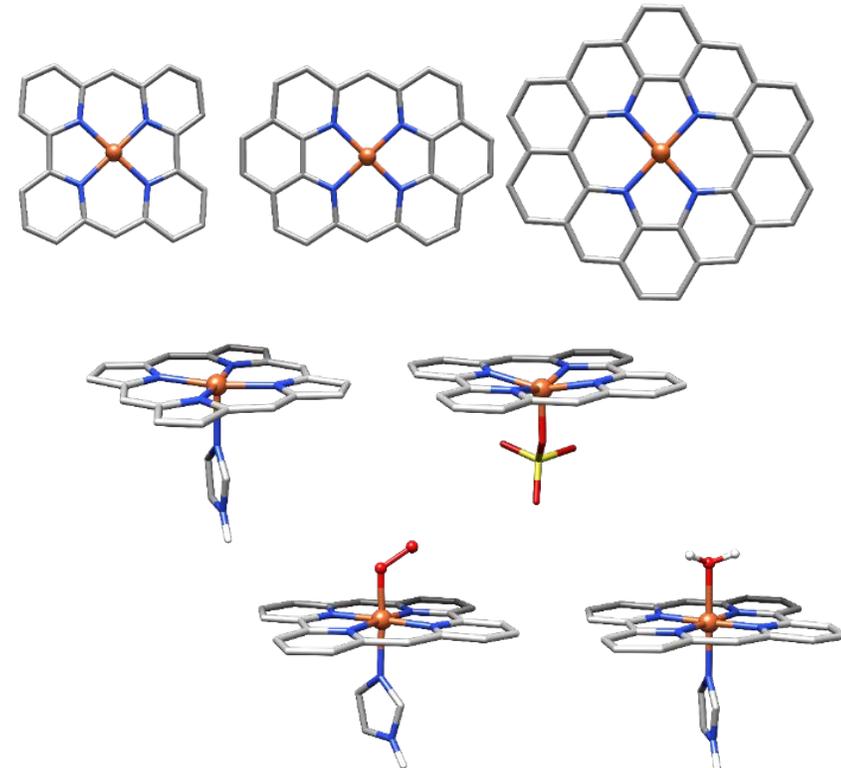
Offene Fragen zum FeNC-Katalysator

Das katalytisch aktive Eisen-Zentrum

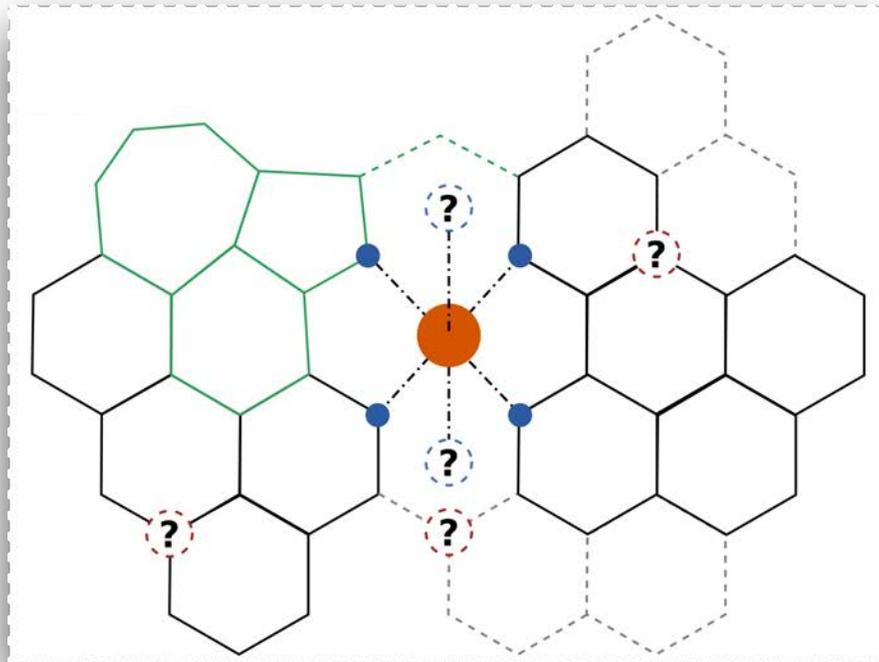
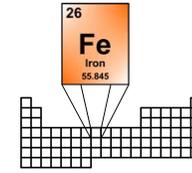


Offene Fragen zum FeNC-Katalysator

(1) Strukturmodelle entwickeln



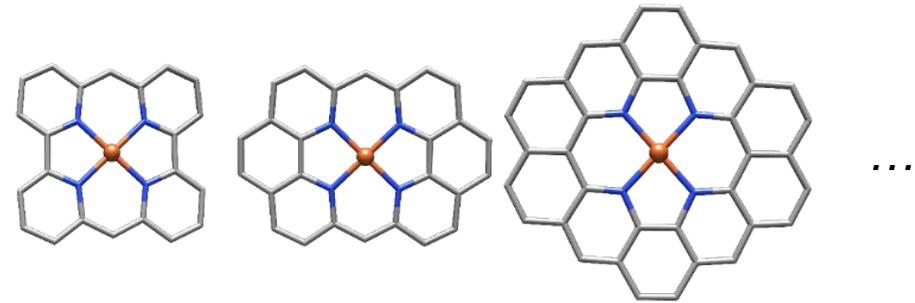
Das katalytisch aktive Eisen-Zentrum



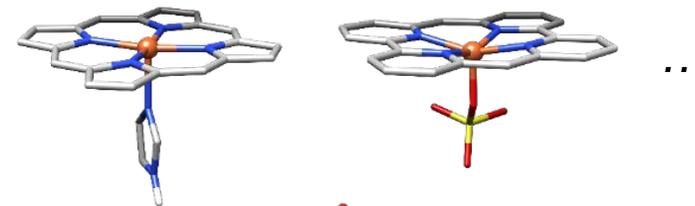
Offene Fragen zum FeNC-Katalysator

(1) Strukturmodelle entwickeln

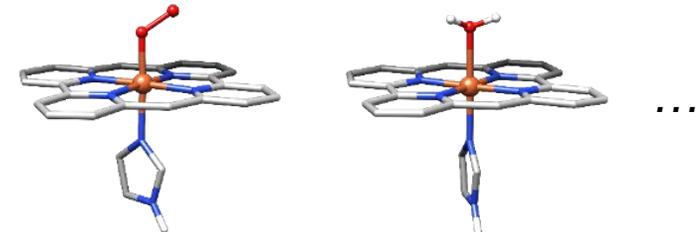
Größe



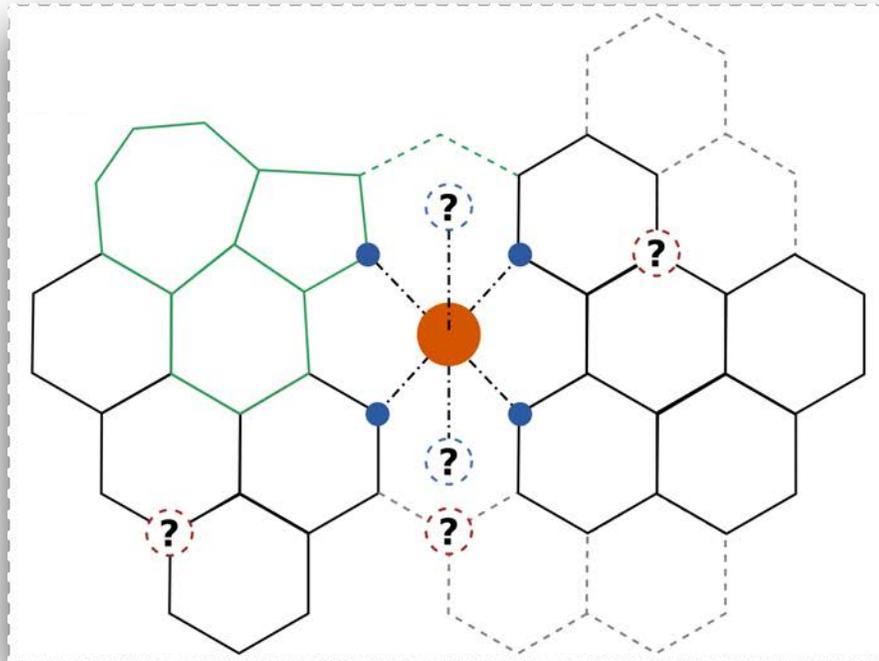
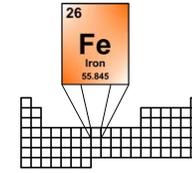
Umgebung



Katalysezyklus

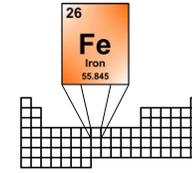


Das katalytisch aktive Eisen-Zentrum

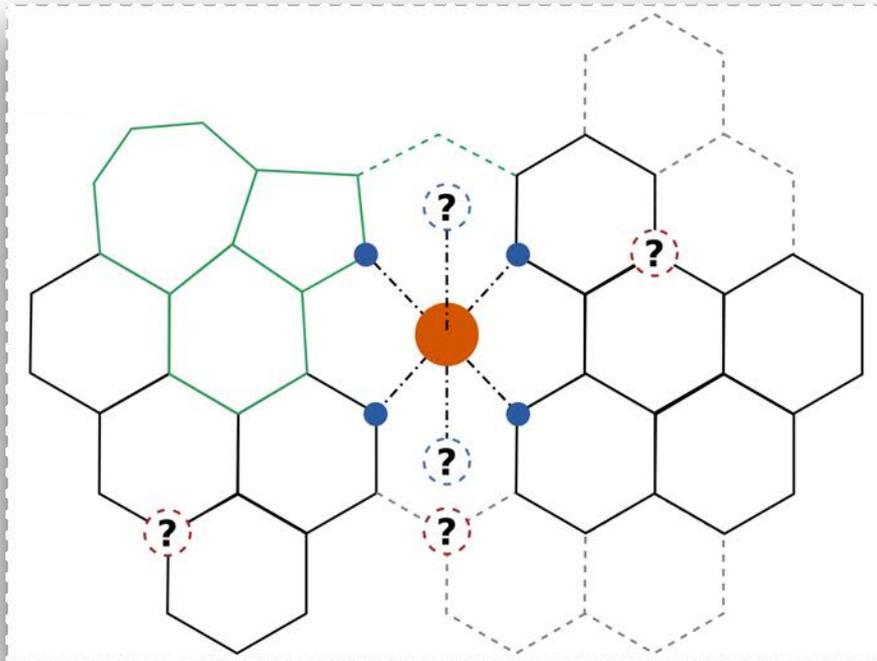


Offene Fragen zum FeNC-Katalysator

Das katalytisch aktive Eisen-Zentrum

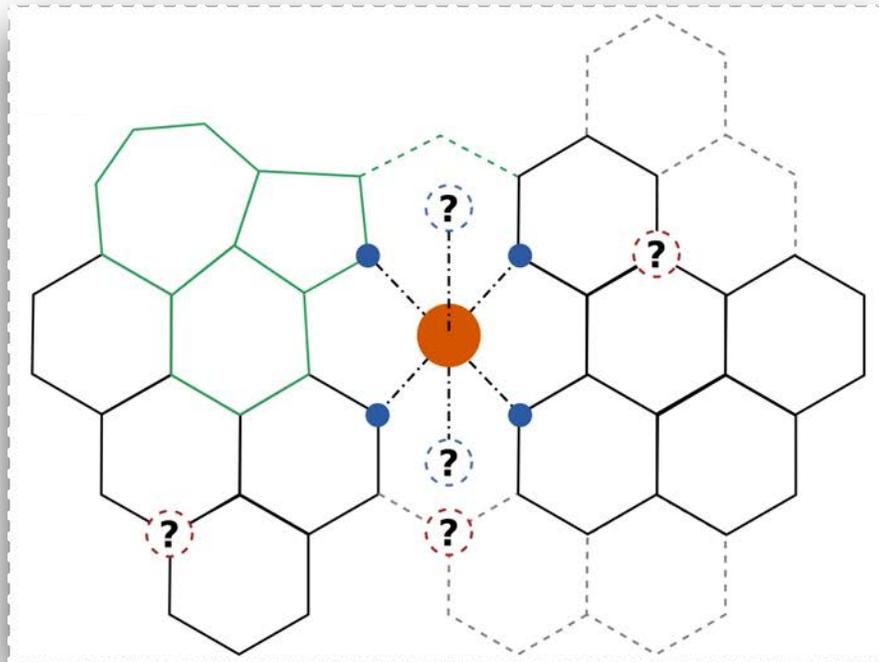
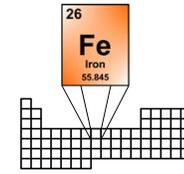


(2) Eigenschaften vorhersagen



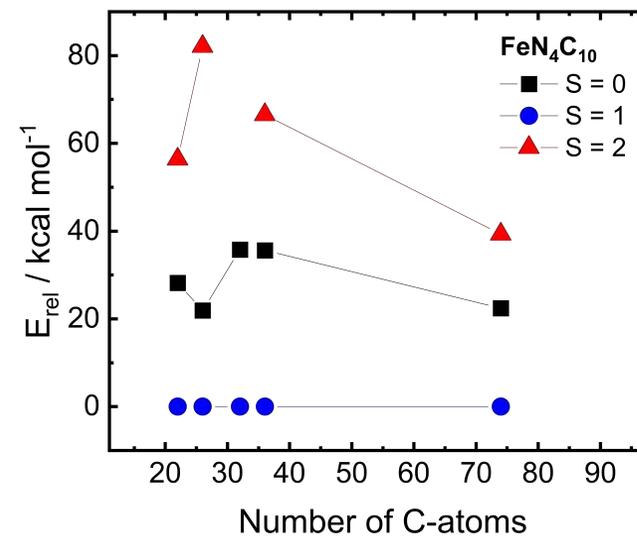
Offene Fragen zum FeNC-Katalysator

Das katalytisch aktive Eisen-Zentrum

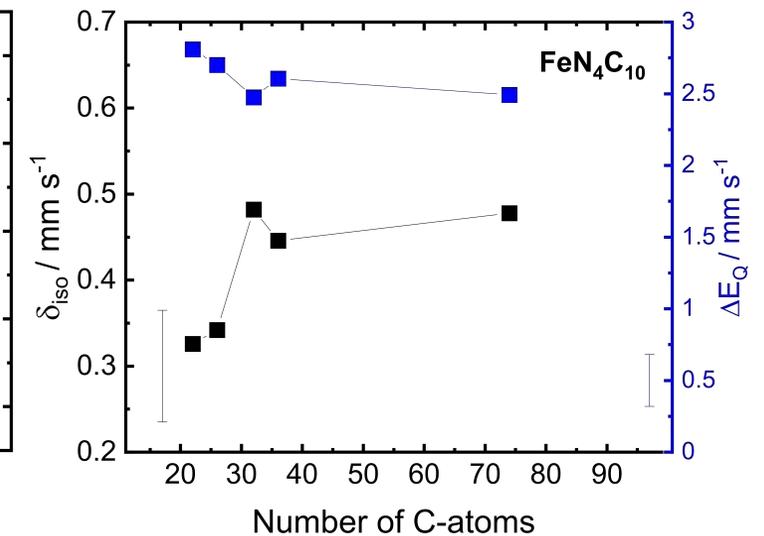


(2) Eigenschaften vorhersagen

Spinzustände

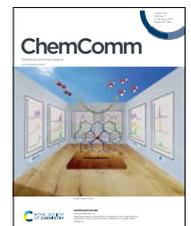


Mößbauer-Spektroskopie

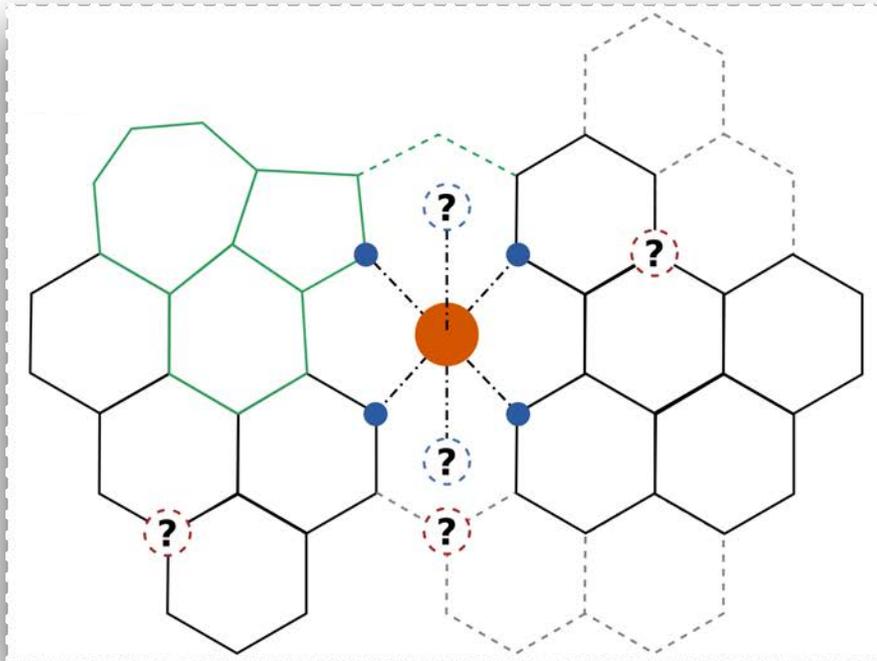
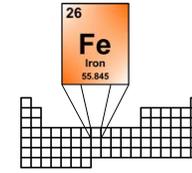


Offene Fragen zum FeNC-Katalysator

Chem. Commun. **2021**; *Adv. Energy Sust. Res.* **2021**; *J. Am. Chem. Soc.* **2022**

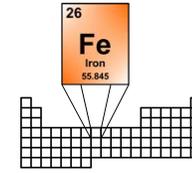


Das katalytisch aktive Eisen-Zentrum

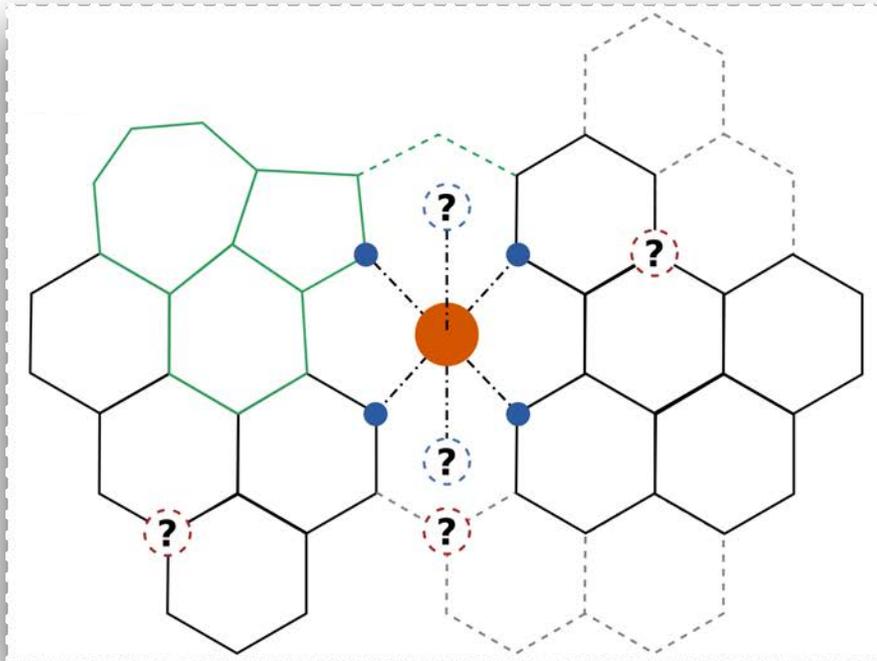


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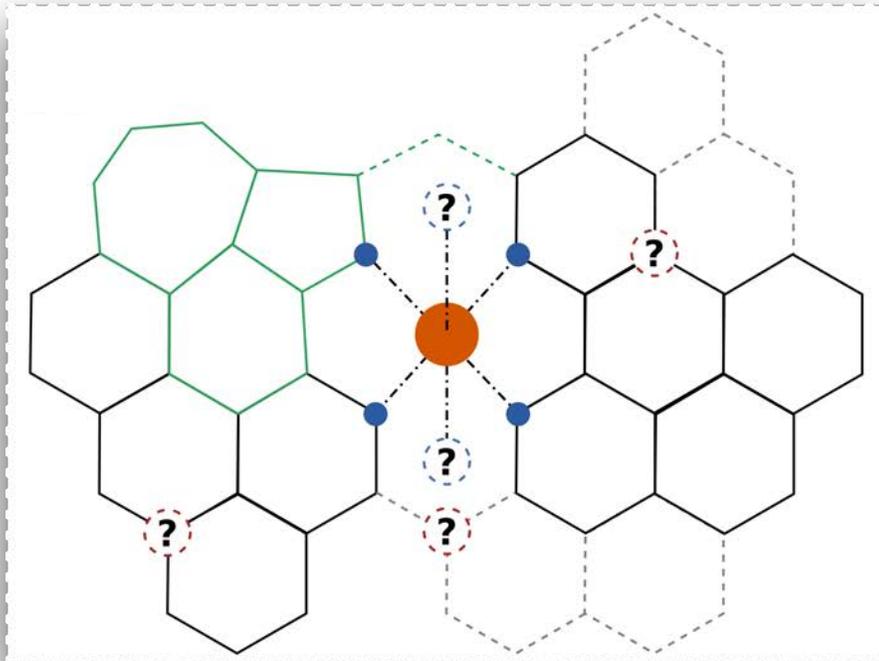
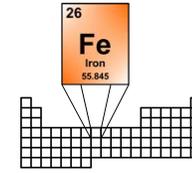


(3) Vergleich mit dem Experiment



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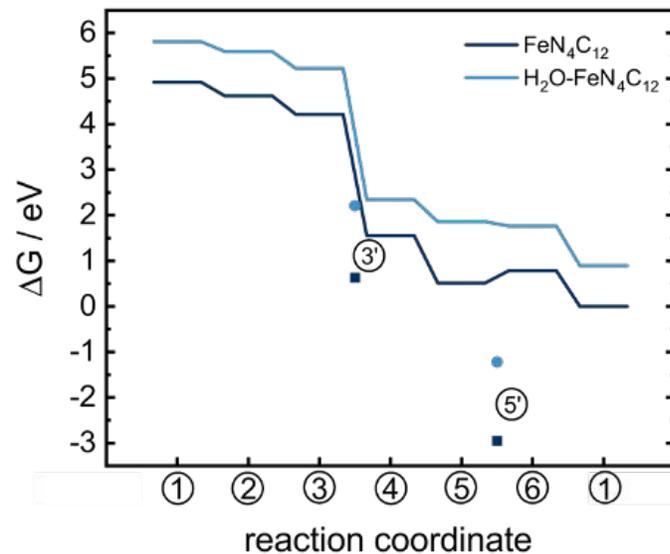
Das katalytisch aktive Eisen-Zentrum



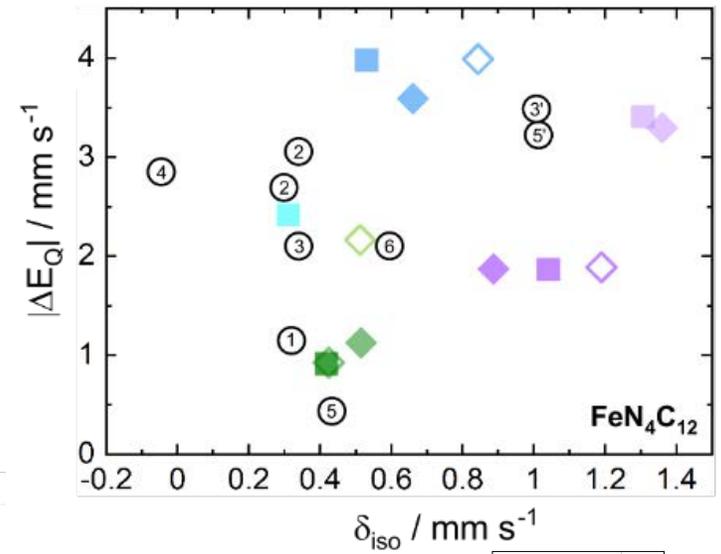
Offene Fragen zum FeNC-Katalysator

(3) Vergleich mit dem Experiment

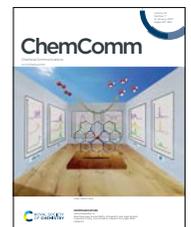
Thermodynamik



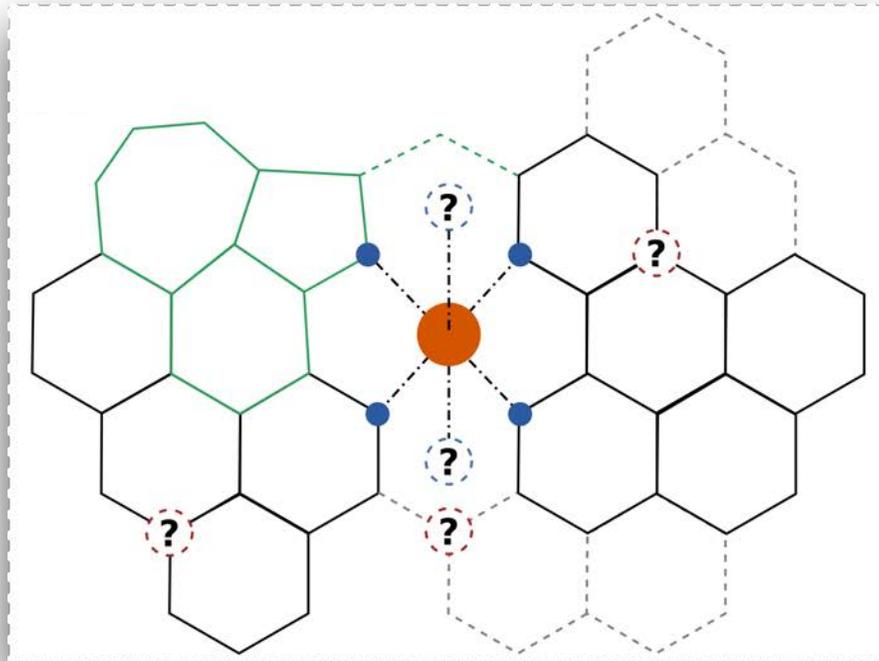
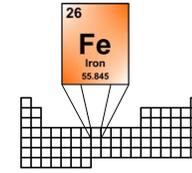
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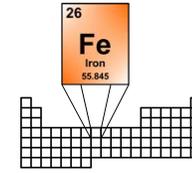


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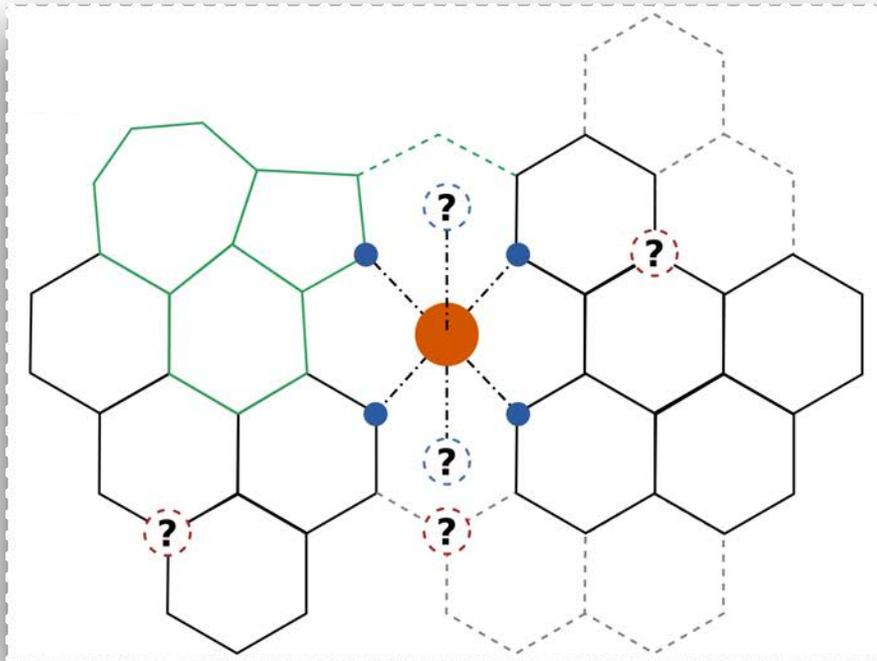


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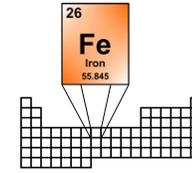


(4) Katalysezyklus

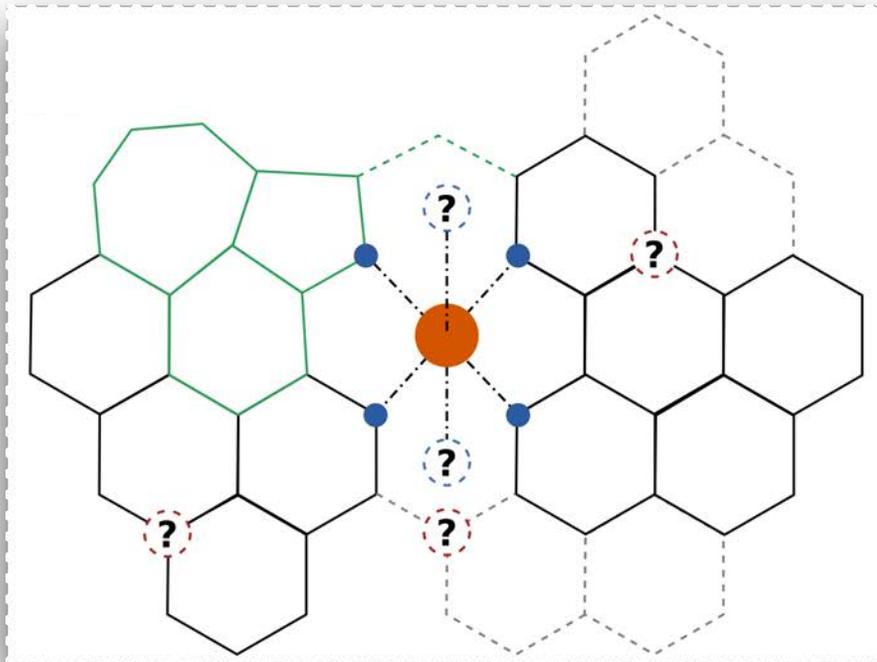
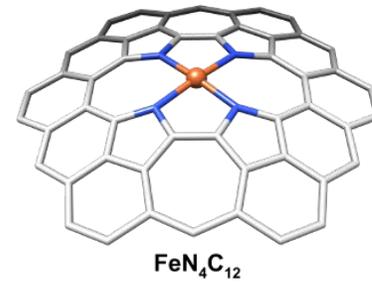


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Das katalytisch aktive Eisen-Zentrum

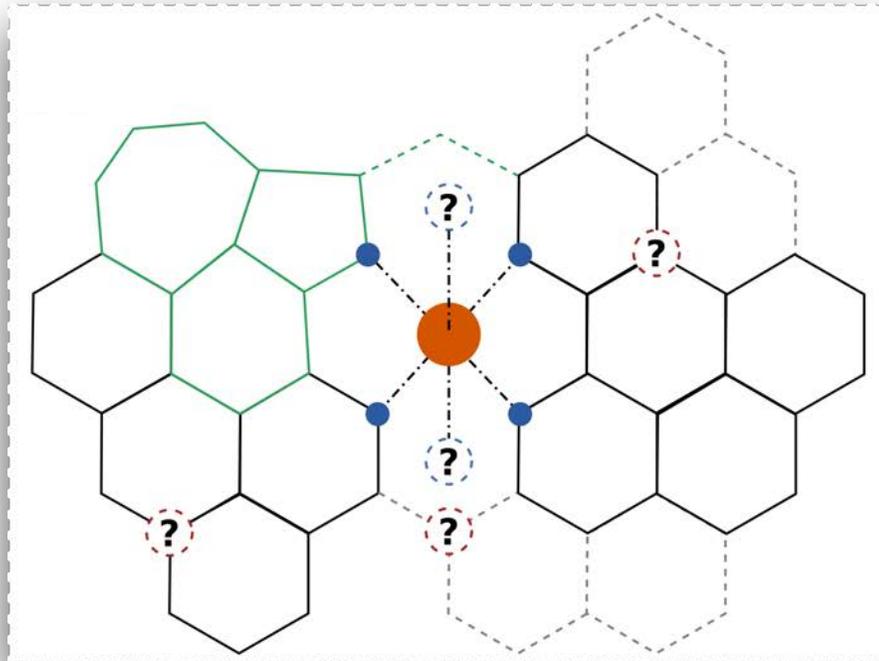
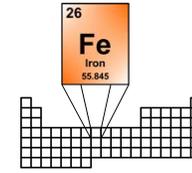


(4) Katalysezyklus



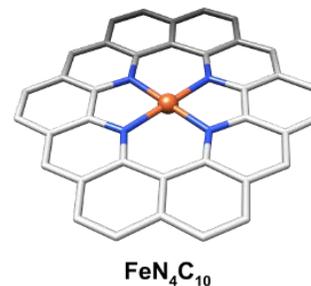
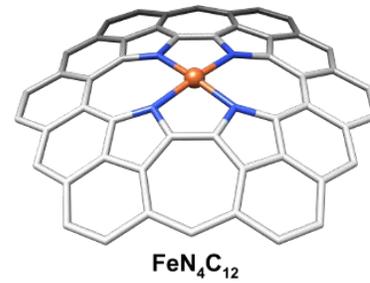
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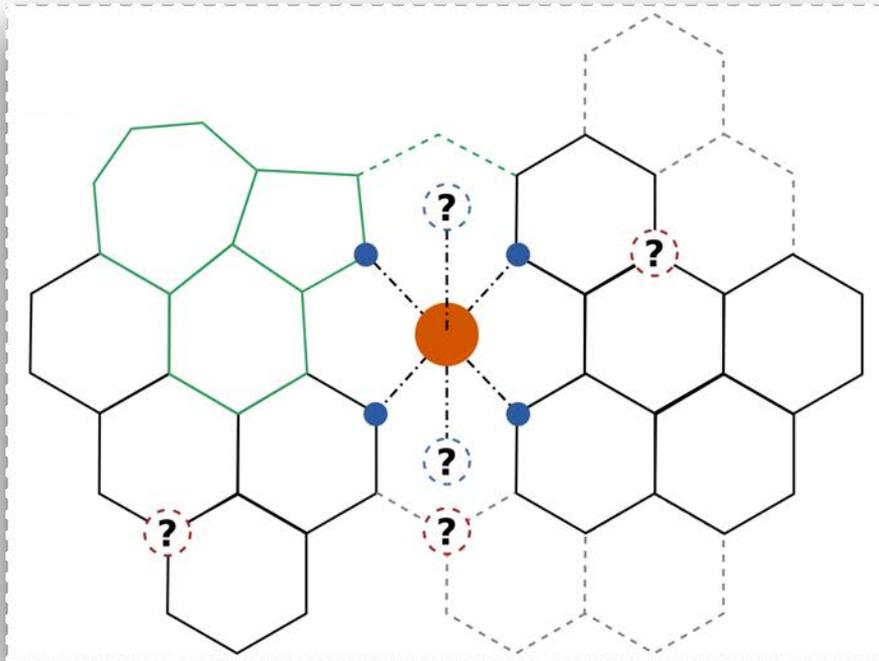
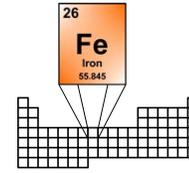


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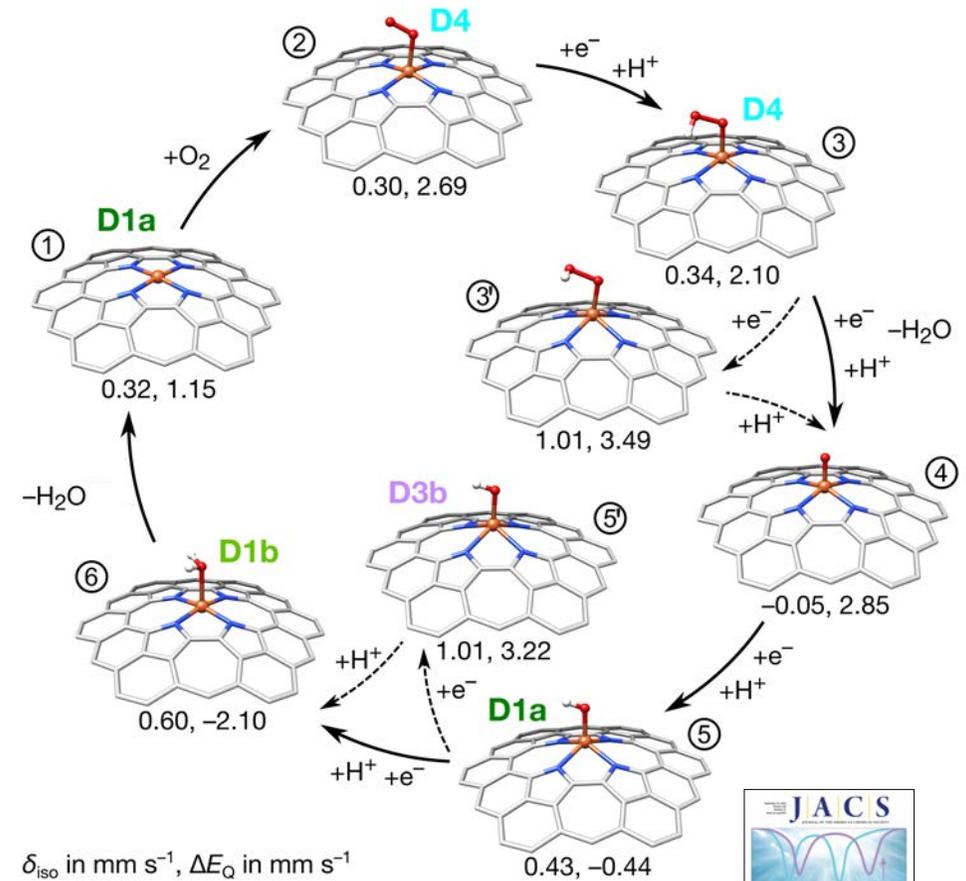
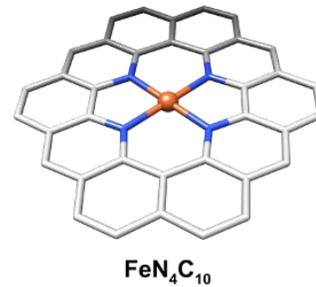
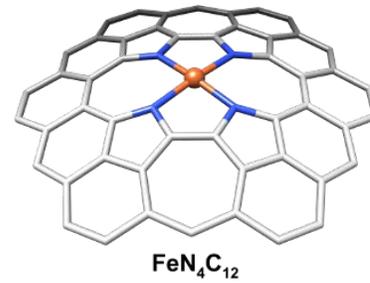


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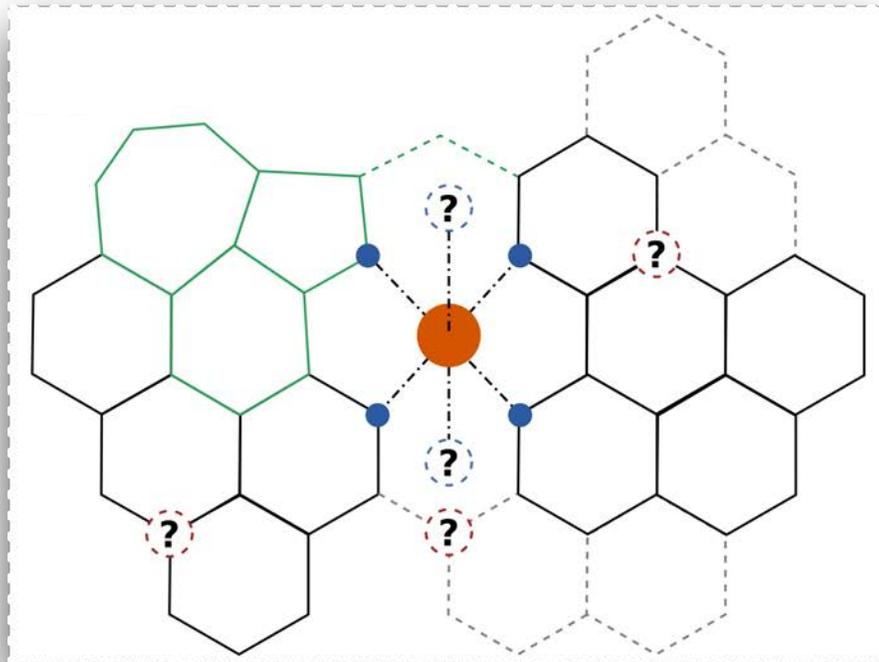
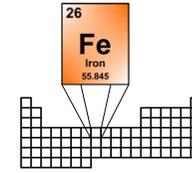
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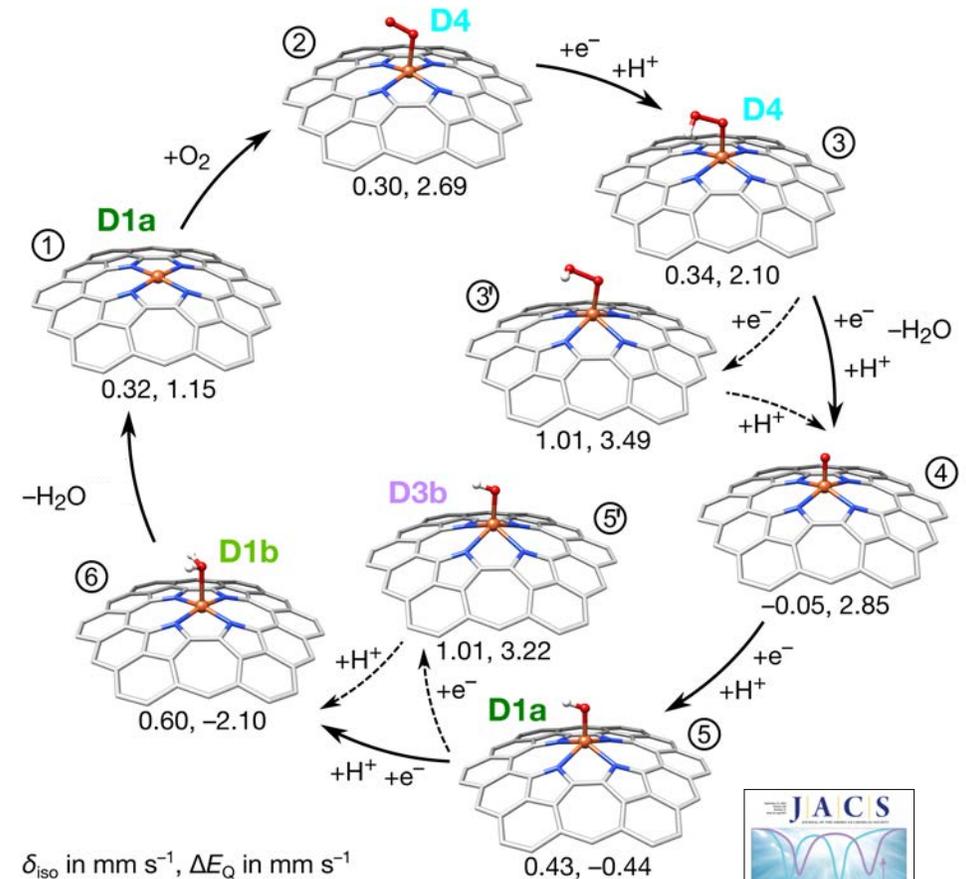
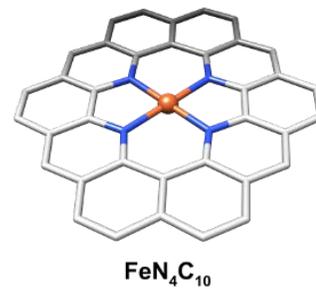
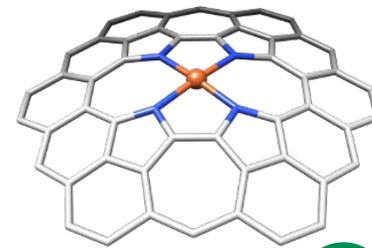


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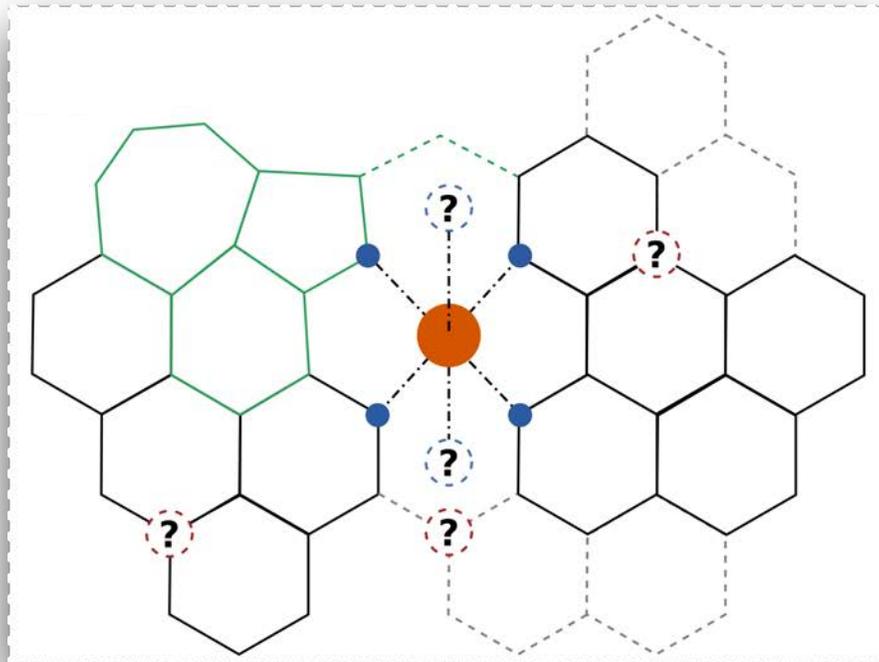
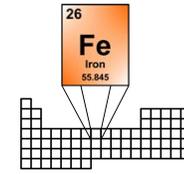
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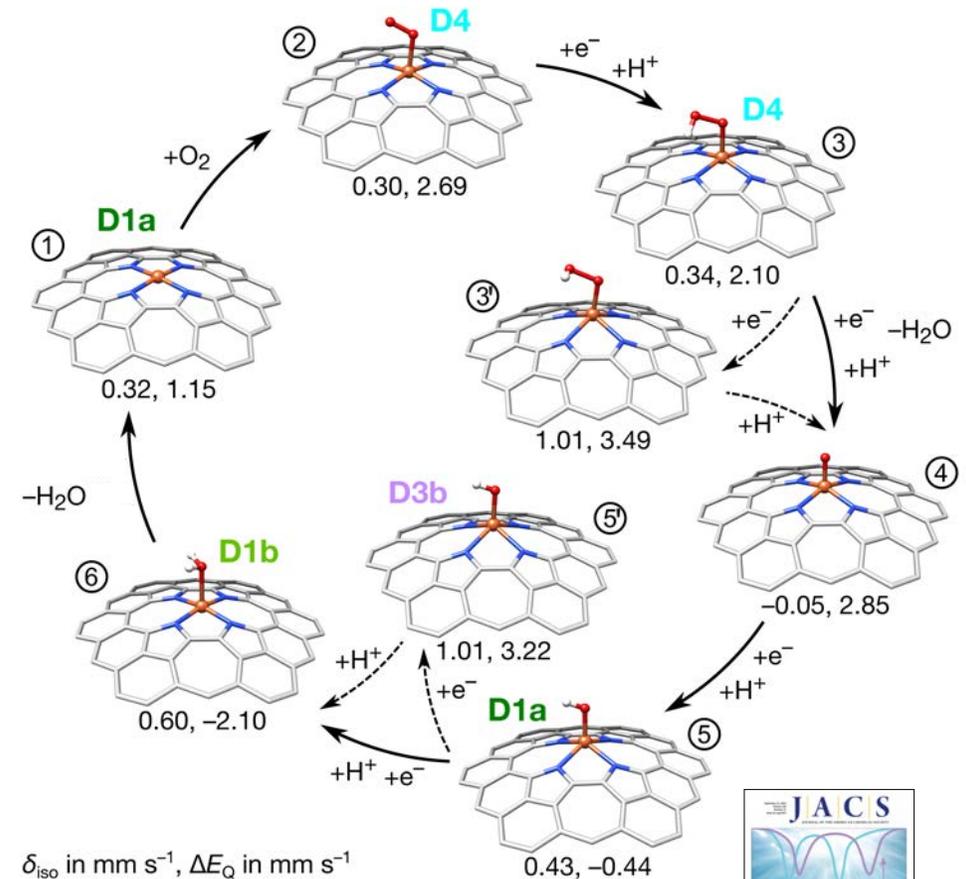
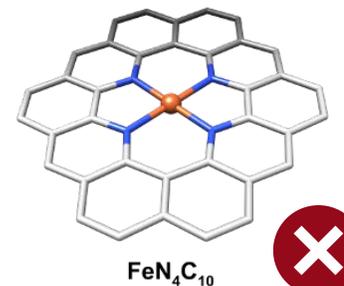
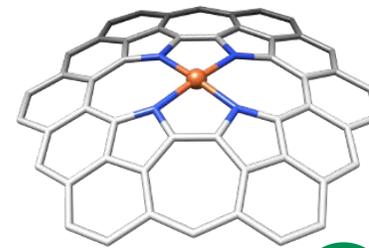


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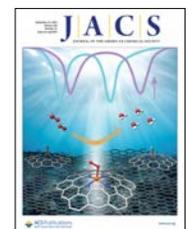


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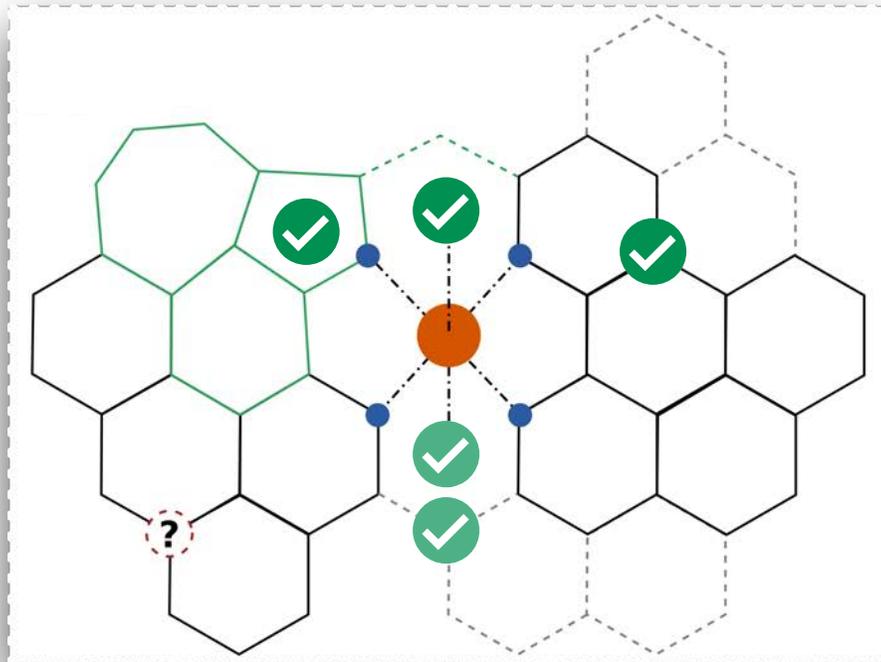
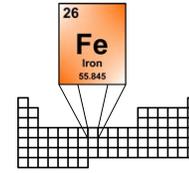
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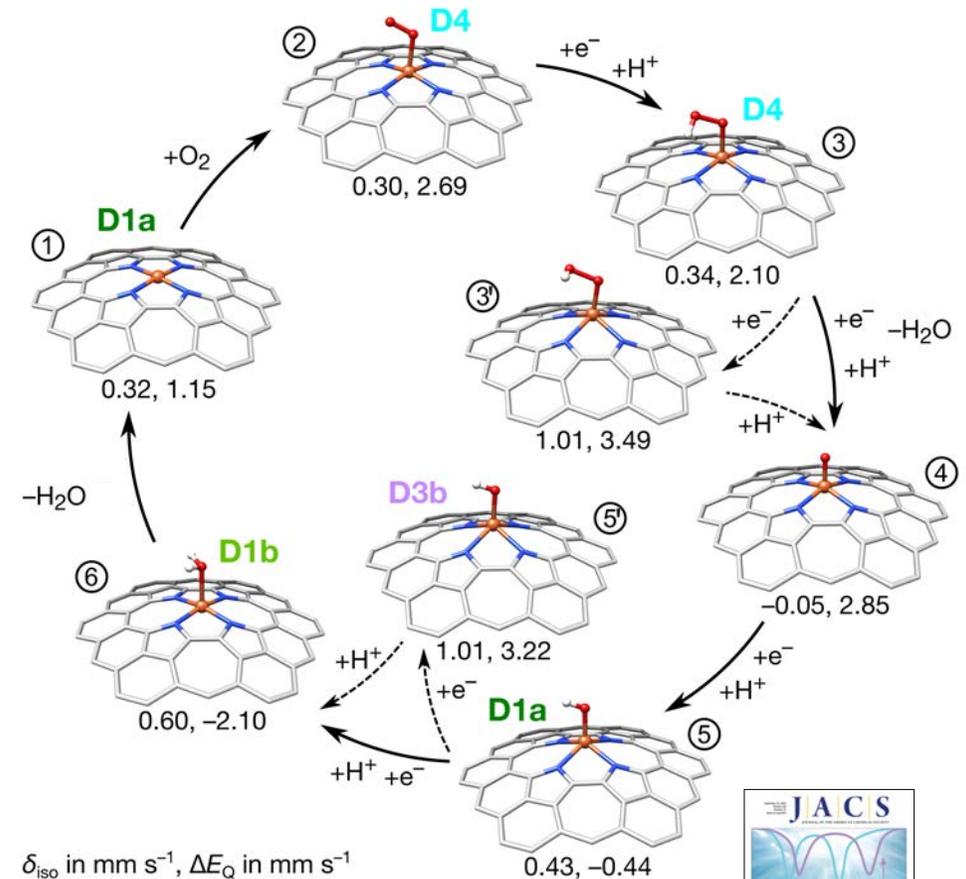
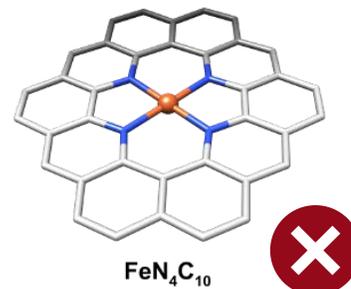
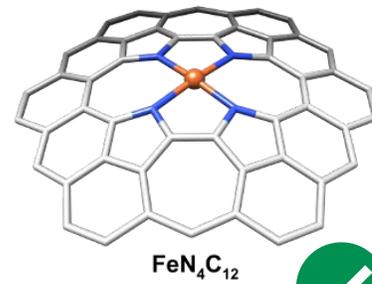


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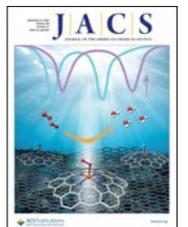


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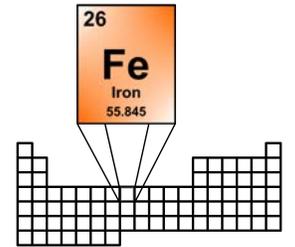
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Quantenchemie für **Katalyse**: nur mit Hochleistungsrechnern!



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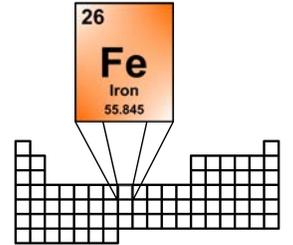
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Quantenchemie für **Katalyse**: nur mit Hochleistungsrechnern!

Katalysatoren eröffnen niederenergetische Reaktionspfade.
Ihre quantenchemische Vorhersage und Analyse ermöglicht
die systematische Verbesserung katalytischer Prozesse.



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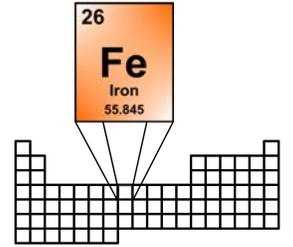
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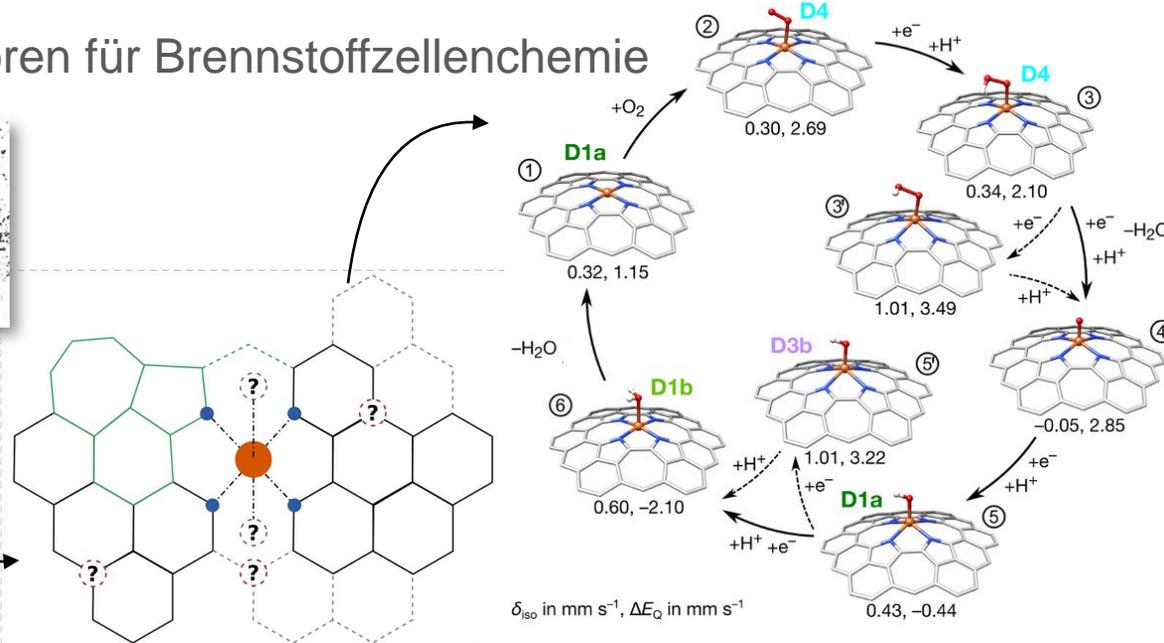


C. Gallenkamp, L. Ni, U. I. Kramm

FeNC-Katalysatoren für Brennstoffzellenchemie



Prof. U. I. Kramm, TUDa



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