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Acid gas valorisation on Na-Faujasite

Catalytic COS formation

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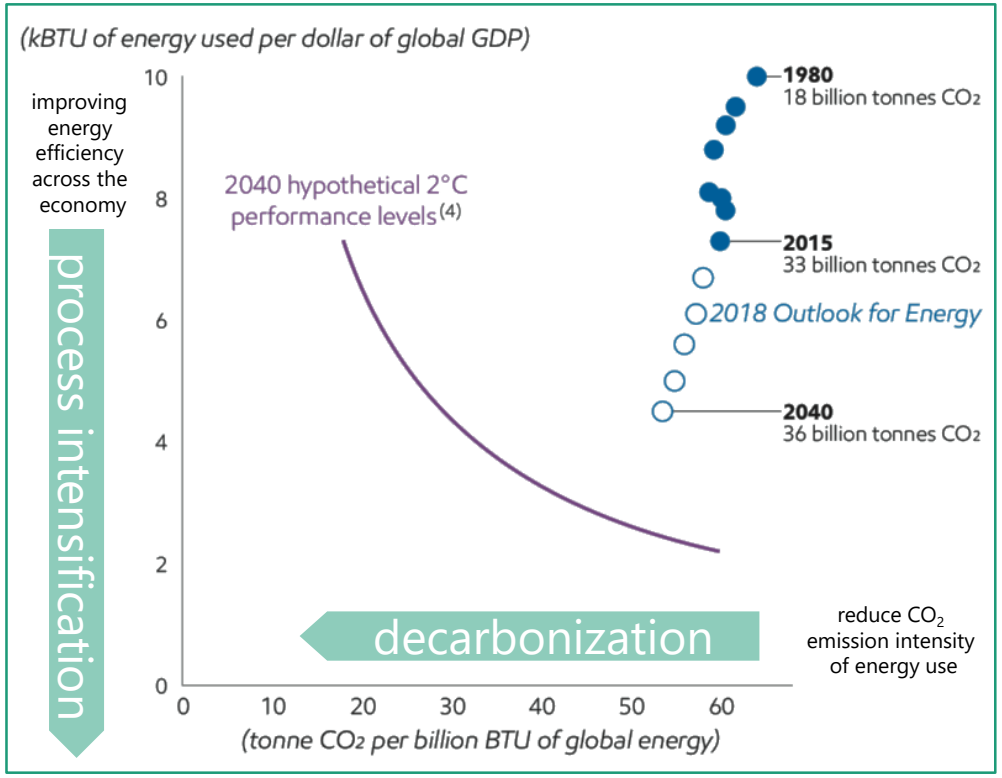
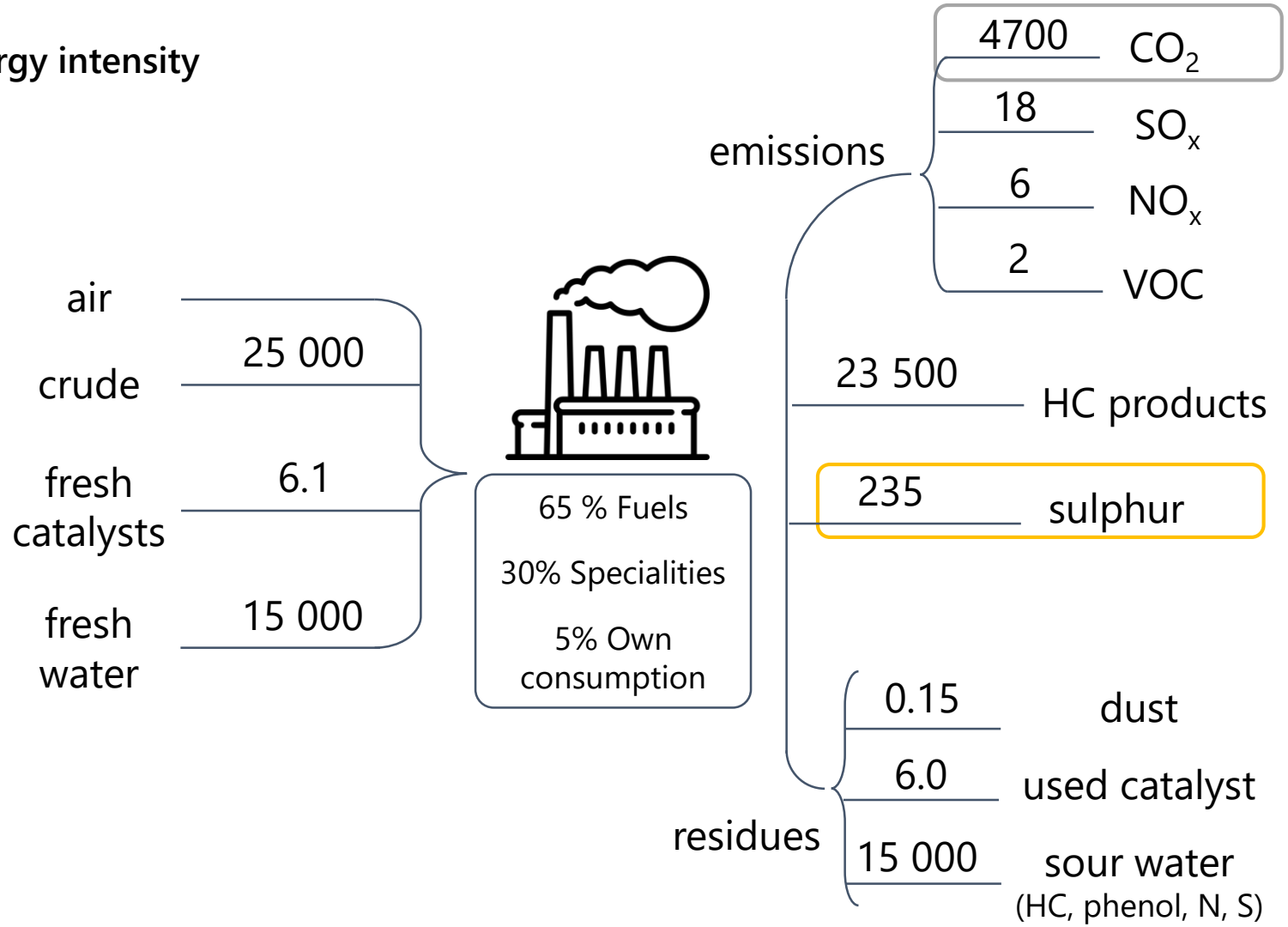
Context : energy outlook

World energy-related CO_2 emissions related to energy intensity vs CO_2 emission intensity

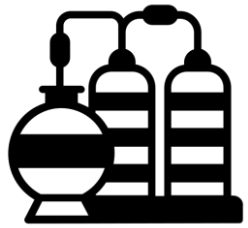


[4] Based on average Stanford EMF27 full technology / 450ppm scenarios' CO_2 emissions (~20 billion tonnes including energy and industrial processes), ExxonMobil GDP assumptions consistent with 2018 *Outlook*



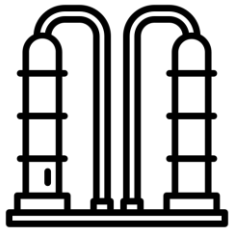
figures are in tonnes per day

Claus vs COS intermediate



Natural Gas

acid gas
$H_2S + CO_2$



separate H_2S and CO_2 streams

mixed CO_2

H_2S (mix)

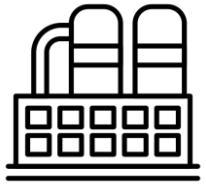
CO_2 purification

pure CO_2

emission

conversion

sequestration



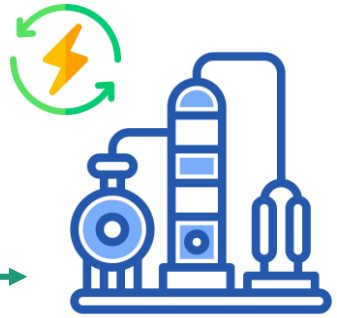
Claus unit

1000°C

$2\ H_2S + 3\ O_2 \rightarrow 2\ SO_2 + 2\ H_2O$
$2\ H_2S + SO_2 \rightarrow 3/2\ S_2 + 2\ H_2O$

recovered sulphur

various stream compositions



e-CODUCT (ETFB)

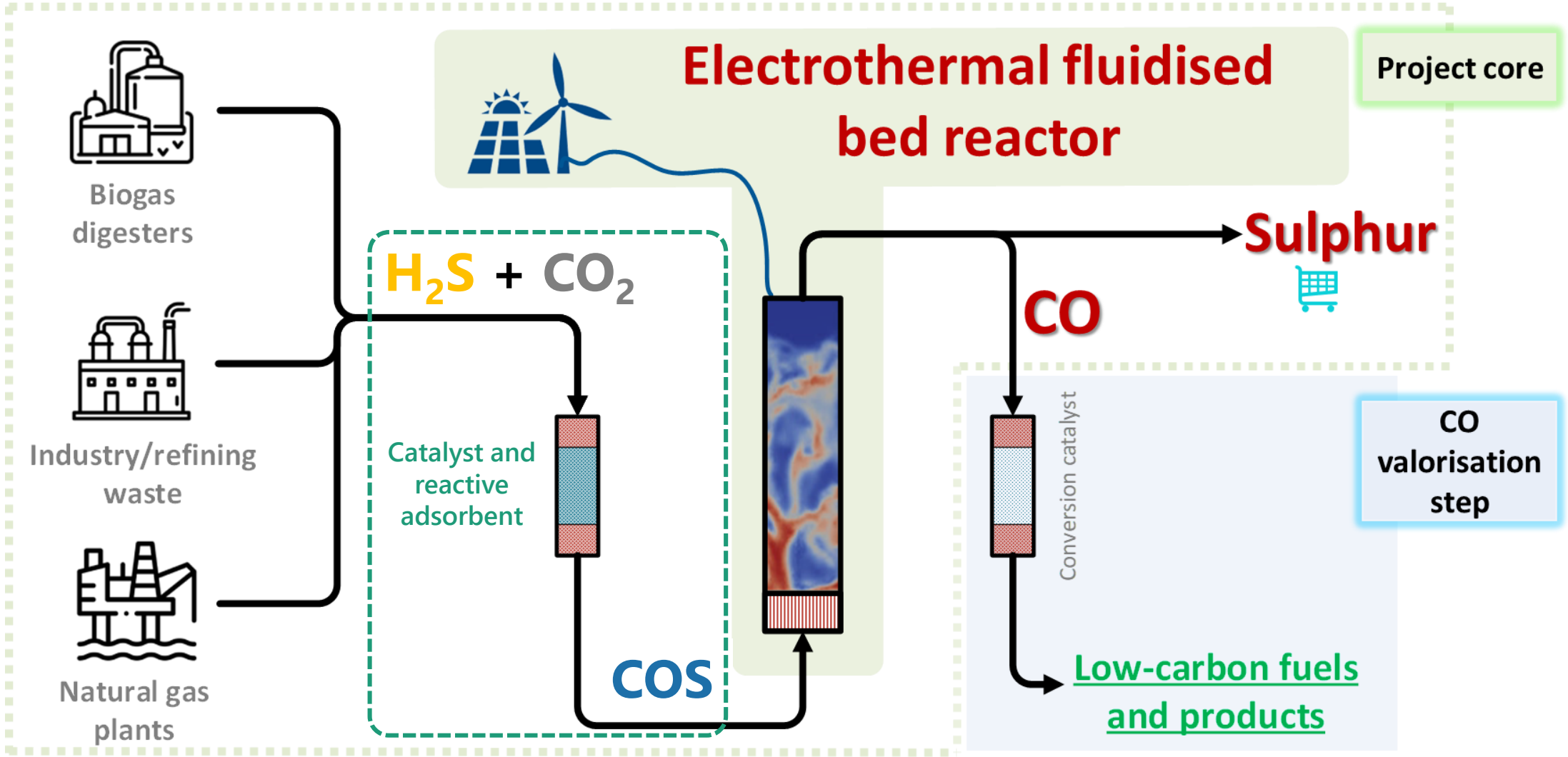
CO_2 → reduced emissions (50%)

pure CO → platform molecule (CH_3OH)

S → marketable

The e-CODUCT process

Horizon Europe
Grant Agreement
N° 101058100



Acid gas conversion on Na-Zeolites

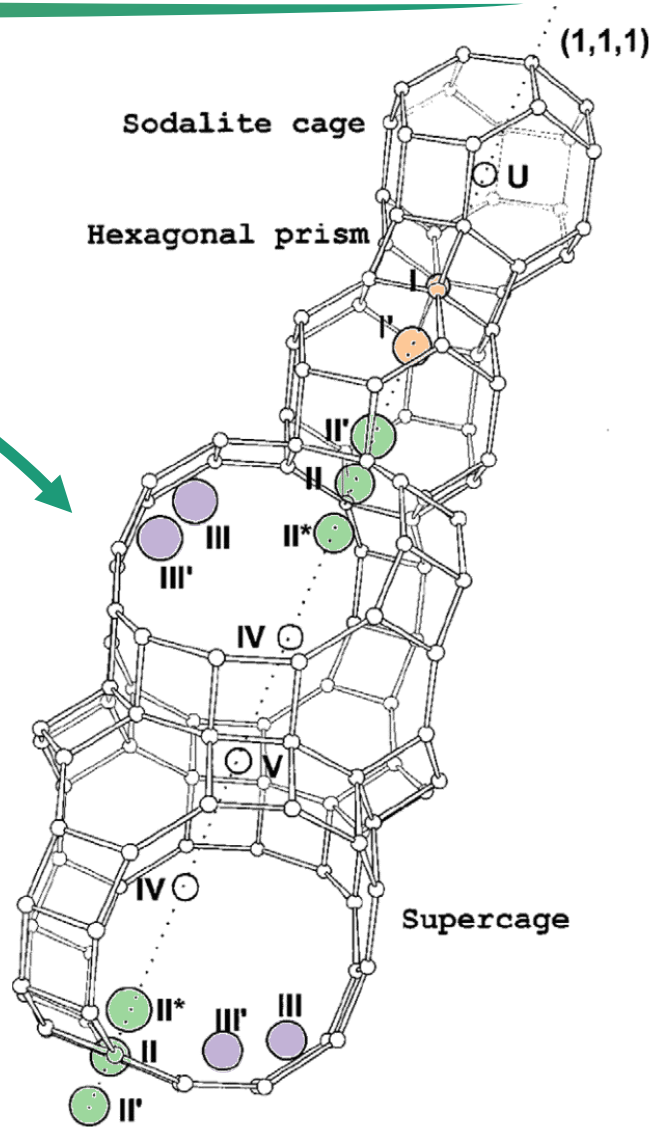
ZEOLITE

$$H_2S_{(g)} + CO_{2\,(g)} \rightleftarrows COS_{(g)} + H_2O_{(x)}$$

Low temperature

Water state (x)	gas	adsorbed
$\Delta_r H^0_{298}$ (kJmol^{-1})	31.0	-11.4
$\Delta_r S^0_{298}$ (JK^{-1}mol^{-1})	0	-118
$\Delta_r G^0_{298}$ (kJmol^{-1})	33.4	23.5
Inversion T (K)	∞	199
K eq. (298K)	1.3E-6	7.5E-5
Pressure impact	0	++

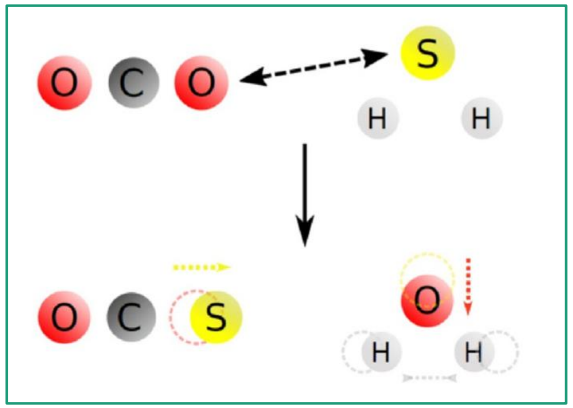
M. Bülow, *Stud. Surf. Sci. Catal.* **1998**



T. Frising, *Microporous Mesoporous Mater.* **2008**

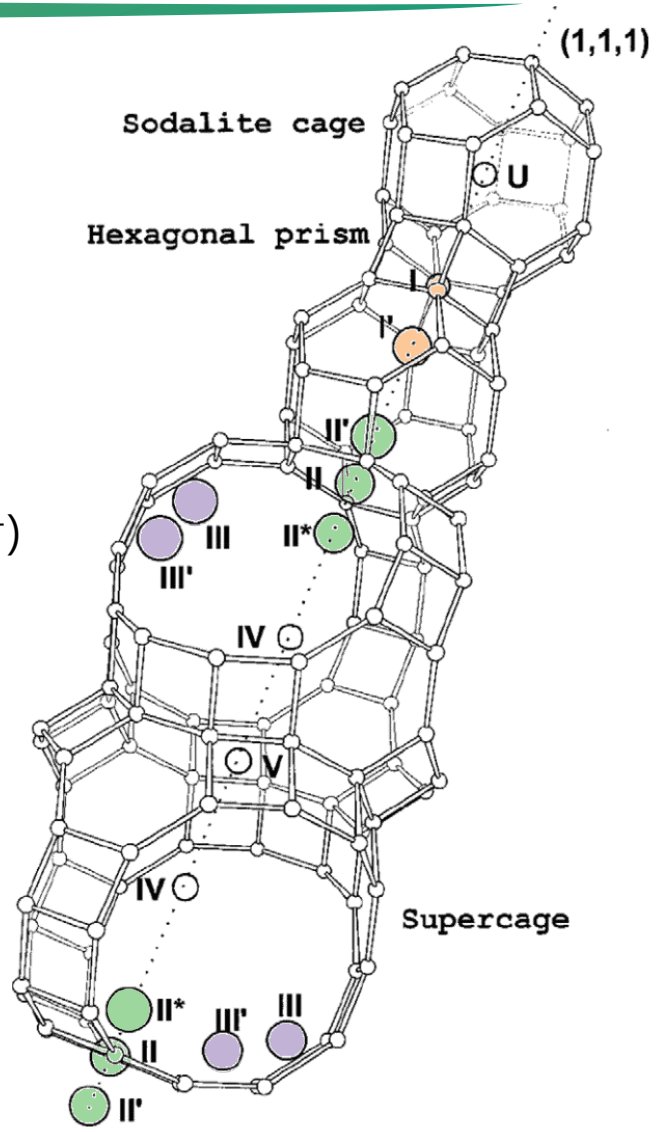
The nature of 13X active site

- Reaction happens on **weakly coordinated Na^+** in the *α cage*
  Fellmuth *Zeolites* **1987**
- *β cages* act as **water sinks** (diffusion controlled, slow kinetics)
  W. Lutz *Adsorpt. Sci. Technol.* **1998**
- In NaY (Si/Al>2.43) **high water activity** in the *α cages* (few low coordinated Na^+)
  Karge **1978**



E. Fetisov, *ChemPhysChem* **2018**

- In NaBEA **Na^+ pairs** yield larger effect on eq. shift than isolated Na
- In NaCaX/A (67-70%) **two sites with different strength** for H_2S dissociative adsorption
  A. Starke *ACSomega* **2022**
- H_2S conversion up to 75%
  (batch experiment, equimolar to Z capacity)
  M. Bülow *Stud. Surf. Sci. Catal.* **1998**



T. Frising, *Microporous Mesoporous Mater.* **2008**

Conclusions

- Catalytic acid gas conversion to COS at mild temperature tested in **mixed feed and with pre-saturation**
- MKM of both sequences, each one requiring a **dedicated set** of parameters
- The amount of **Na cations accessible to H_2S** is used as the total amount of active sites
- Catalyst **deactivation due to water poisoning** active sites, where *sod* do not participate in water placement even for long reaction times
- Optimization: effect of **temperature**, pressure, etc.

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Thankyou for the kind attention

Merci pour l'attention