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Sonohydrothermal synthesis of zeolite A and its phase transformation to sodalite

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MONTPELLIER

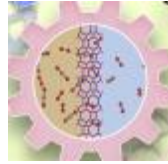


Zeolites are crystalline aluminosilicates (and related compositions) with framework structures containing well-defined micropores

gas separation



ion exchange



catalysis



environmental protection



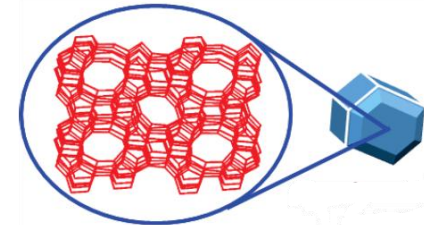
Conventional zeolite synthesis



calcination



zeolite



(Si, Al, Ti....) precursors
Structure directing agents
Mineralizers (OH^- or F^-)

Amorphous hydrogel

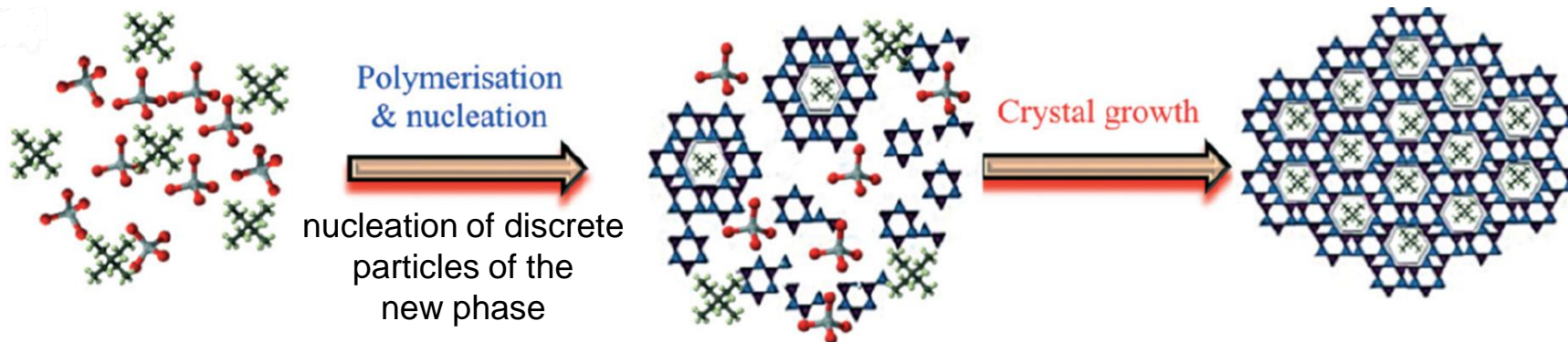
Hydrothermal heating

- ◆ Mixing of the raw materials to give a precursor (gel) mixture
- ◆ **Hydrothermal treatment** at elevated temperature for different times (**hours to days**)

Zeolite synthesis



The crystallization of zeolite proceeds via two steps



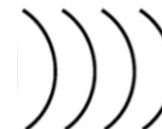
Hydrothermal synthesis
(HT)



Time & Energy consuming process

(because of the required restructuring)

New alternative synthesis route



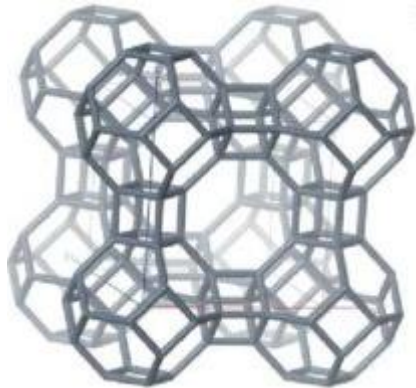
Simultaneous coupling of HT conditions with power ultrasound

⇒ **Sonohydrothermal (SHT) synthesis**

Structure of zeolite A

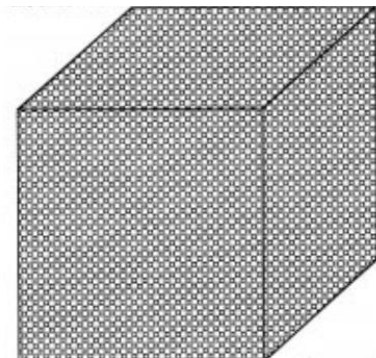


The structure of zeolite A consists of a **cubic array of cages forming a 3D network**



Zeolite A structure

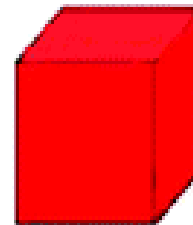
Sodalite structure



Cubic zeolite A crystal

conversion
→

Dissolution / Precipitation



Sodalite crystals

Metastable phase

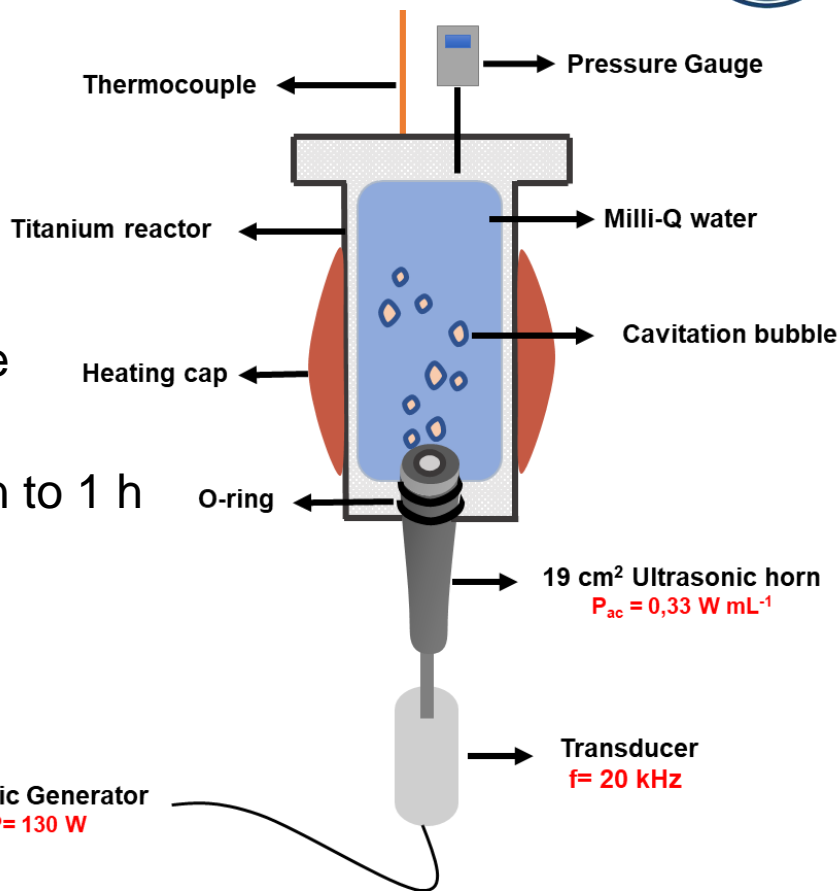
Stable phase

Hydrothermal (silent conditions) versus Sonohydrothermal (SHT) synthesis of zeolite A



SHT

Frequency: 20 kHz
Temperature: 100 °C
Autogeneous pressure
Volume: 50 mL
Synthesis time: 15 min to 1 h



Gel preparation

SiO_2 : 1,6g } 21 mL H₂O
 NaOH : 2,1g }

NaAlO_2 : 4,7g } 27mL H₂O

- ♦ Mechanical stirring (**MS**)
30 min

or

- ♦ 20 kHz **US**, 70% amplitude,
30 min

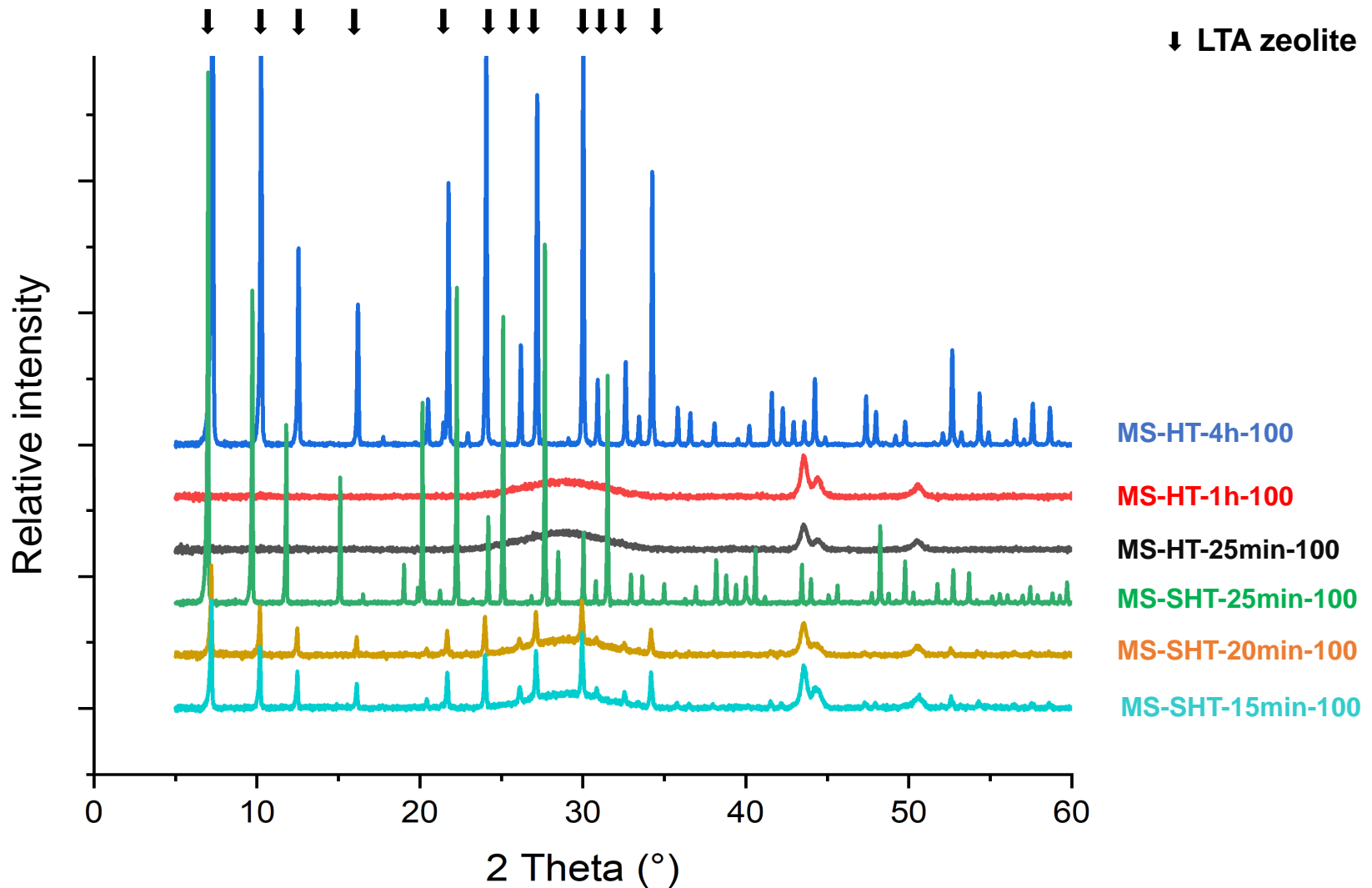
HT (silent conditions)

Parr reactor

100 °C to 150 °C, 25 min to 48 h



SHT vs. HT: influence on the crystallization of zeolite A

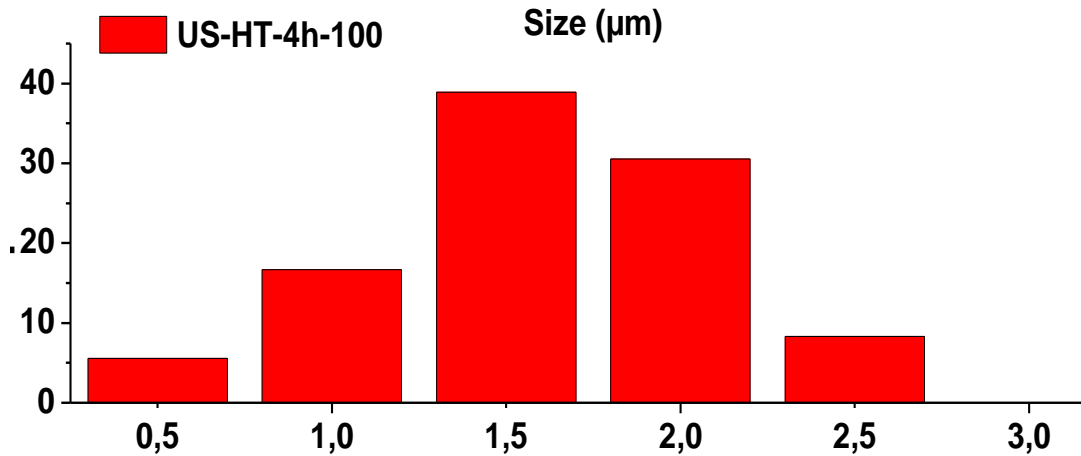


Conversion of the amorphous gel to zeolite A in the SHT reactor is 9.6 times faster than in hydrothermal conditions

Effect on crystal size - Hydrothermal synthesis

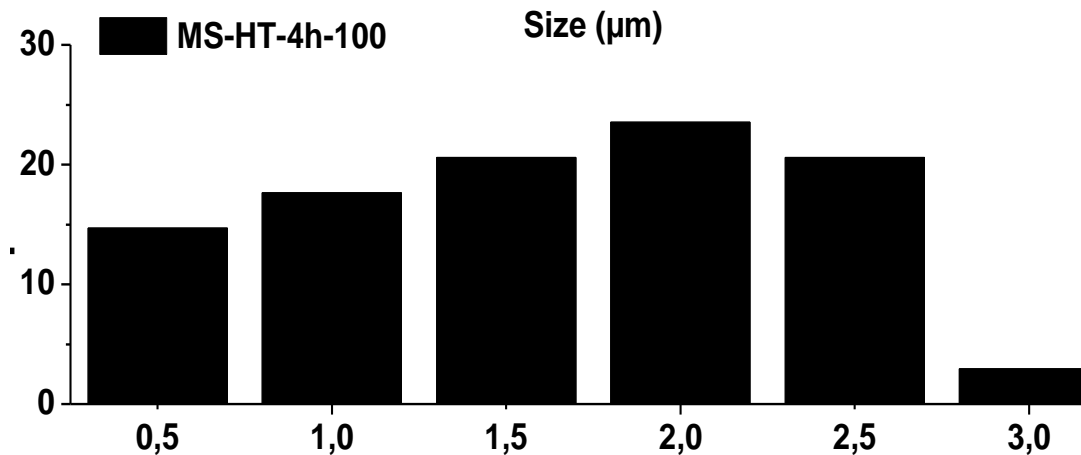


Crystal size calculated using ImageJ software



Method used for the hydrogel preparation

US: ultrasonic precipitation



MS: mechanical stirring

More homogeneous, centered particle size distribution for the solid based on ultrasonic precipitation prior to the heating treatment (**US-HT-4h-100**)

Effect on crystal size - SHT synthesis



Crystal size calculated using ImageJ software

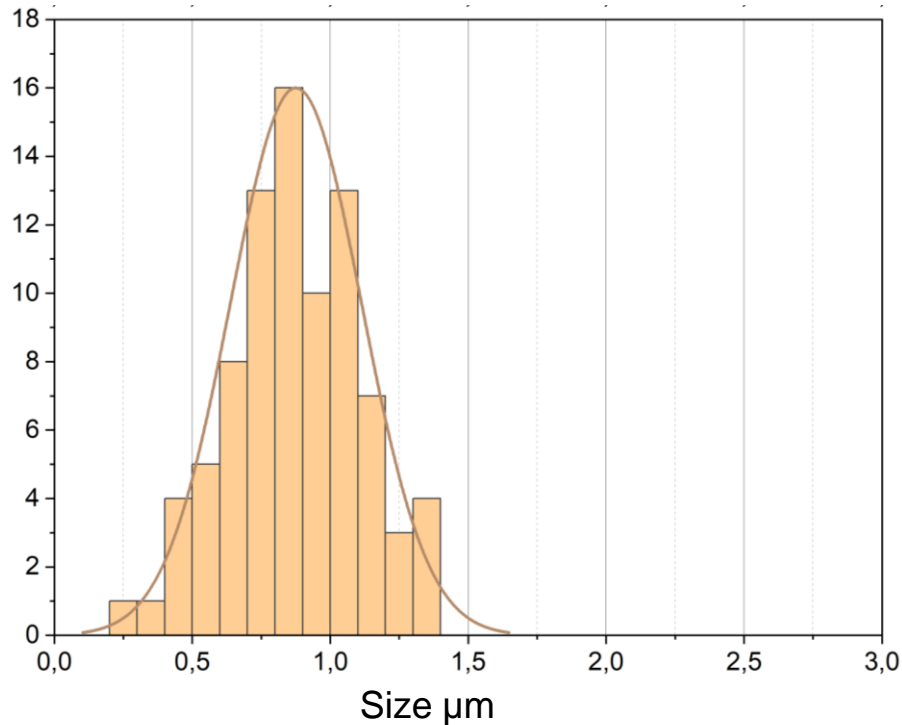
Method used for the hydrogel preparation



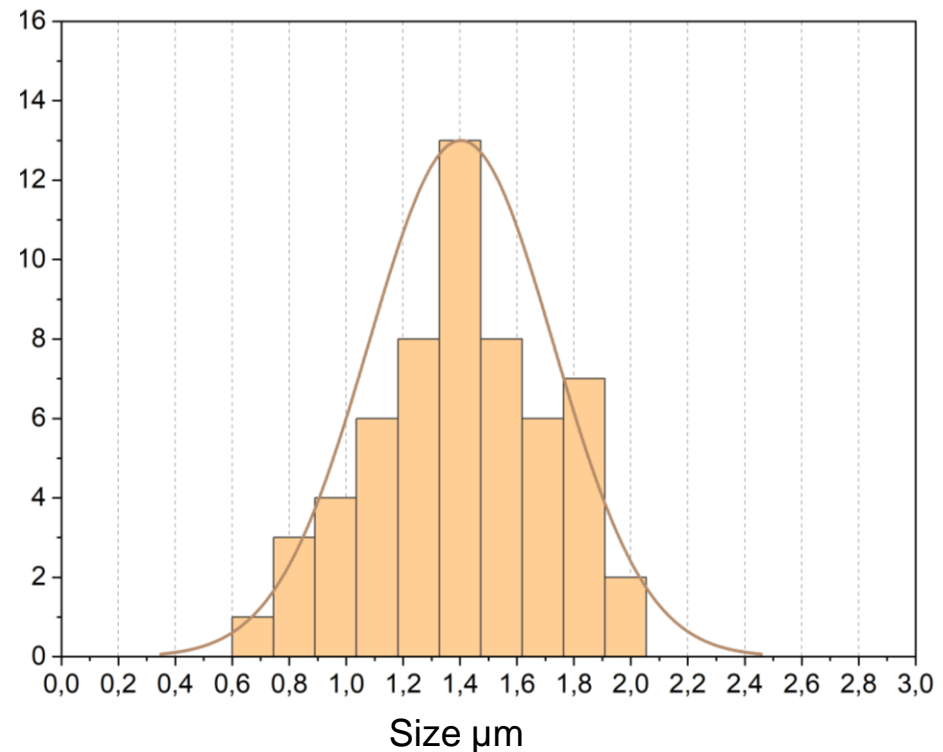
US: ultrasonic precipitation



MS: mechanical stirring



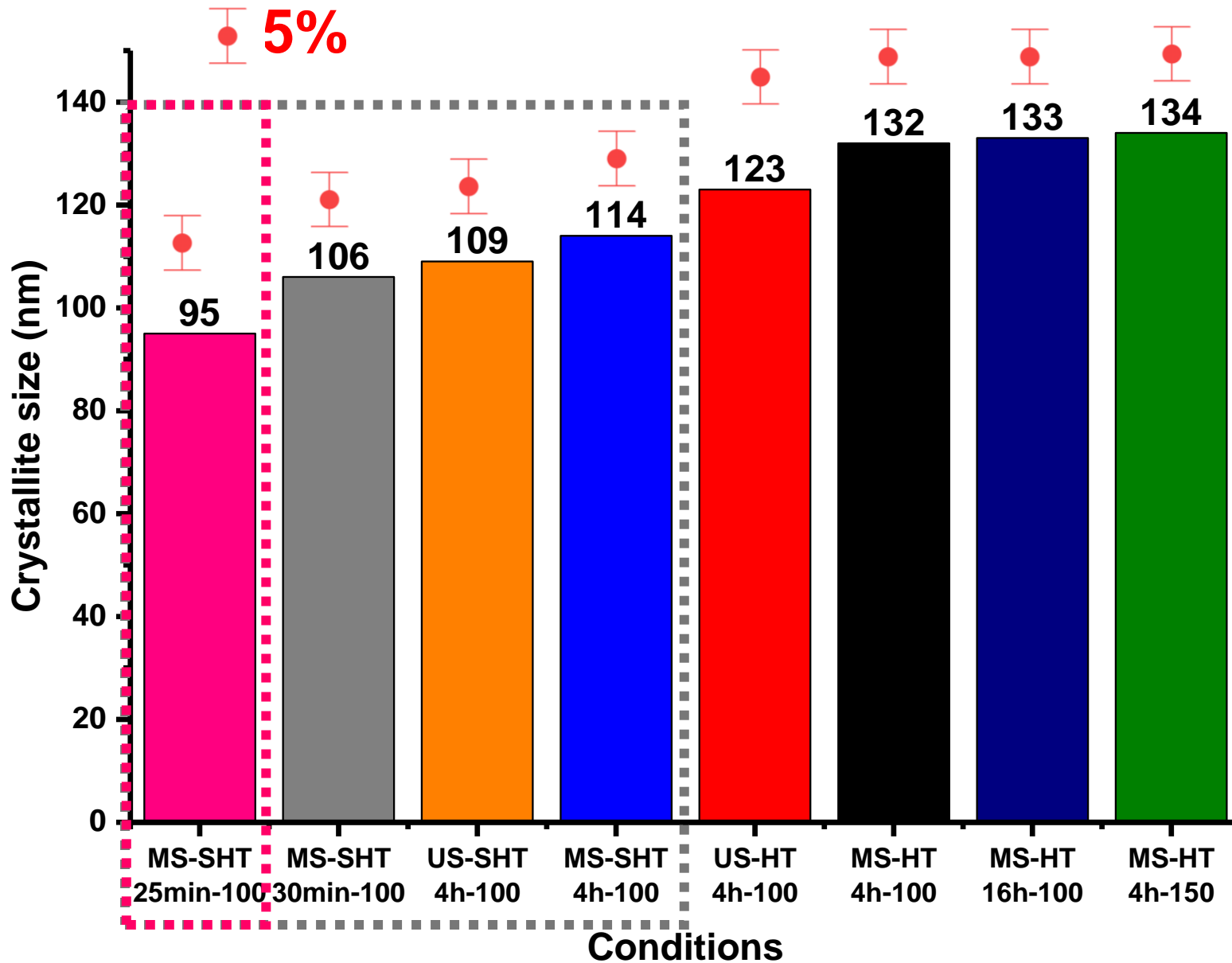
US-SHT-100 °C-1h



MS-SHT-100 °C-1h

Smaller crystals size for the specimen based on ultrasonic precipitation prior to the SHT treatment

Zeolite A crystallite size evolution

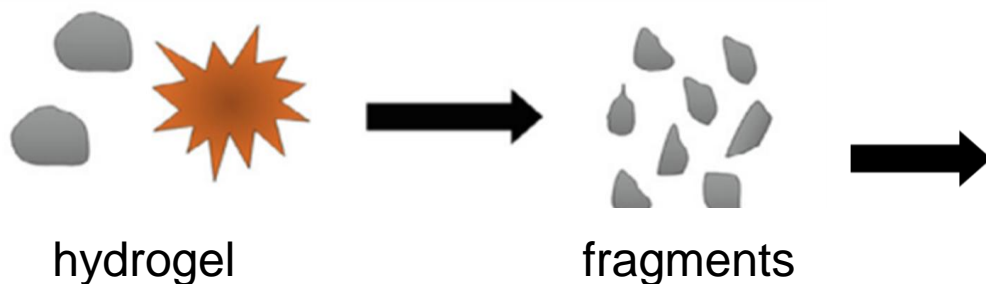


Crystallite size determined using the Scherrer formula

Sonofragmentation

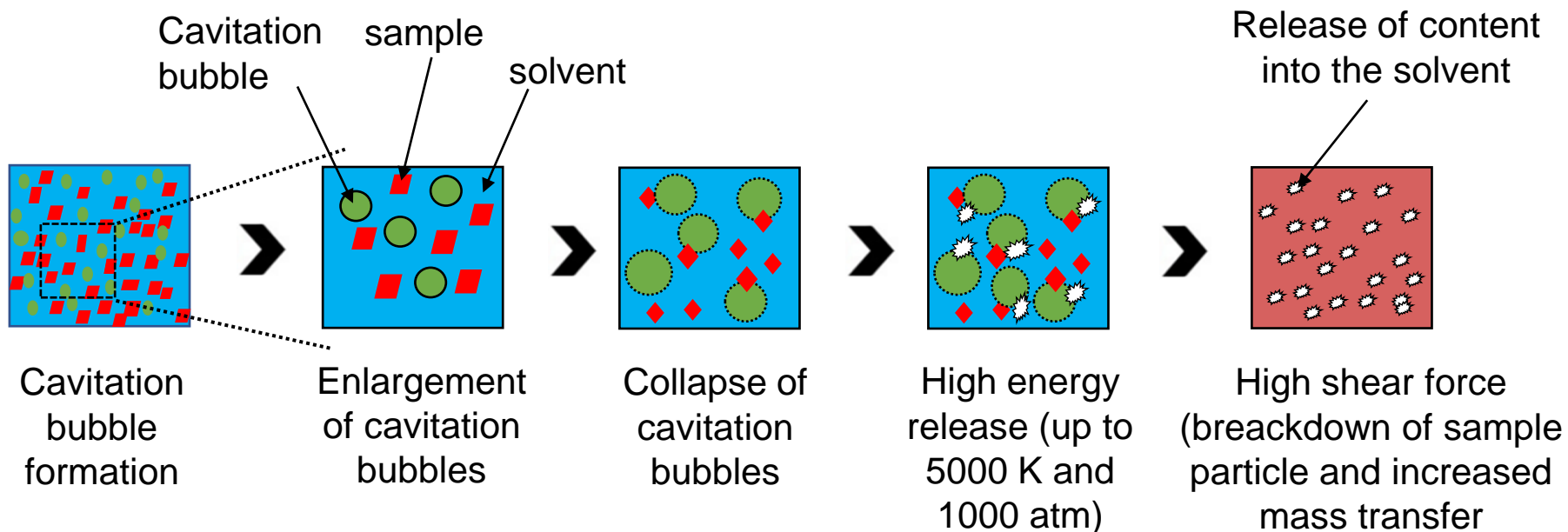


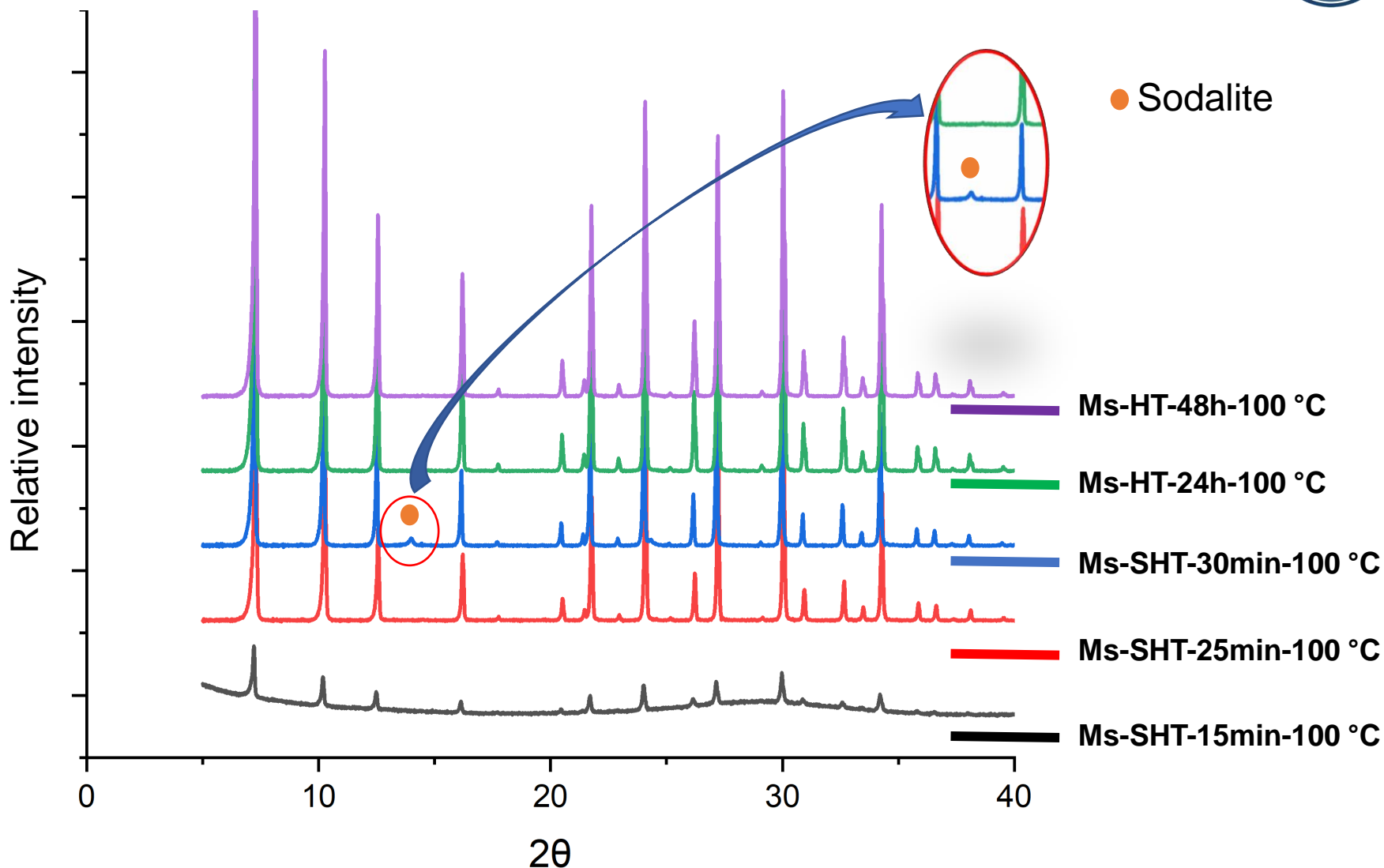
Gel mixture (precursor)



SHT conditions at 20 kHz
⇒ shockwave effects

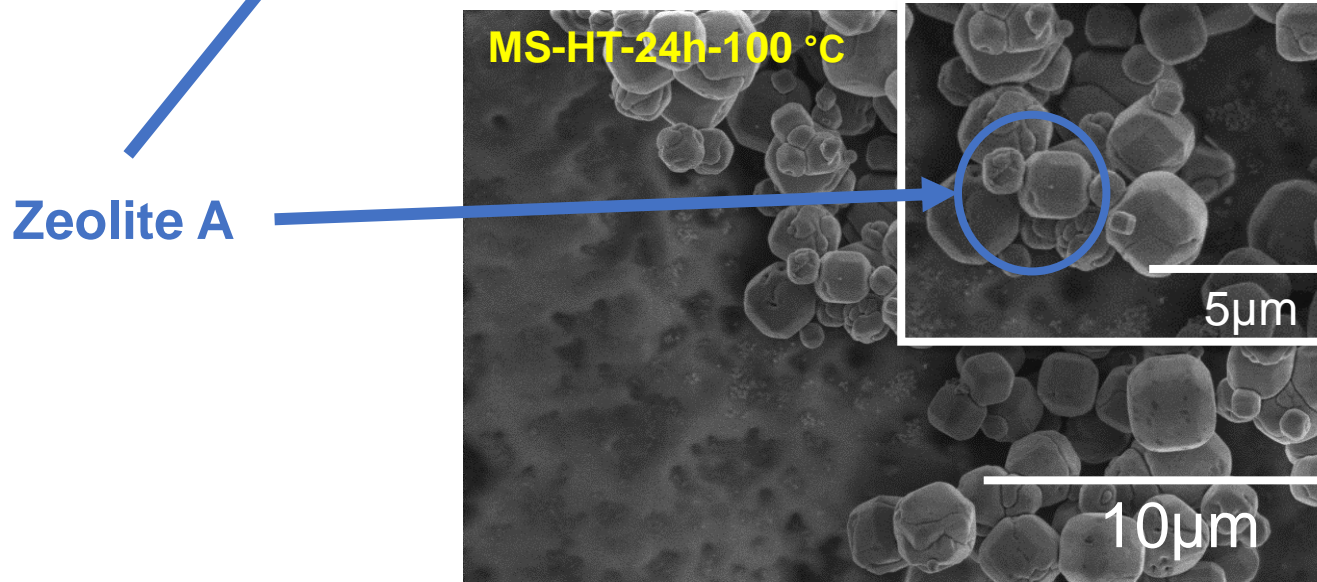
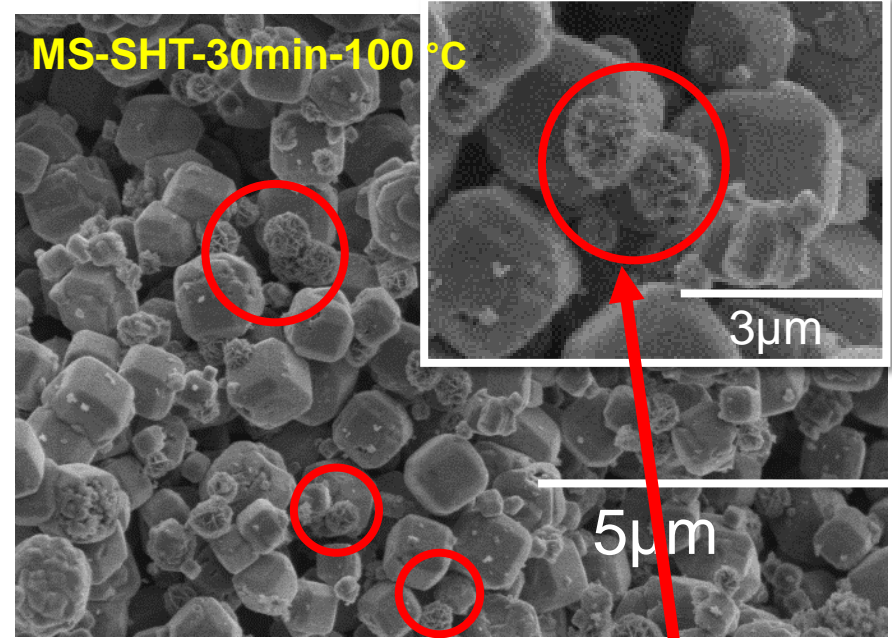
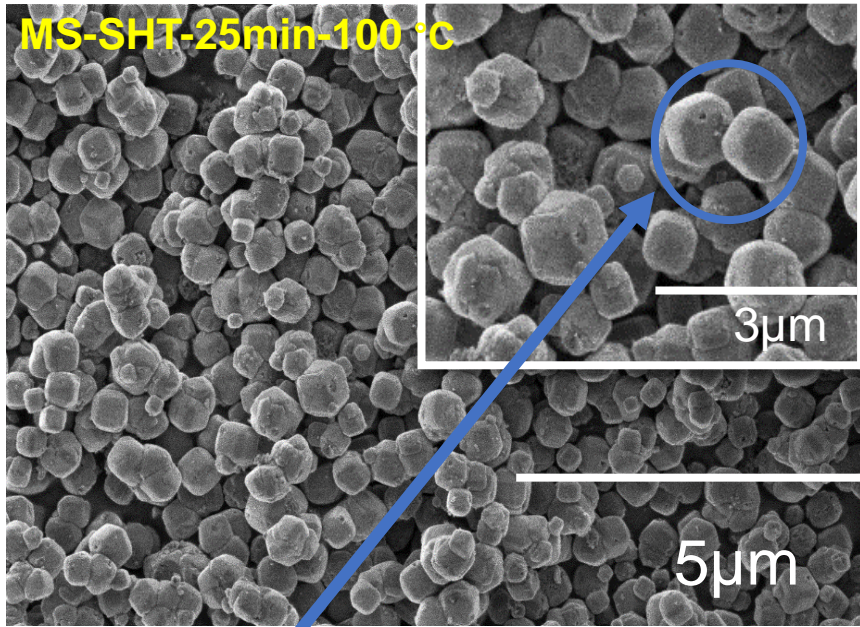
- Gel fragmentation
- More nucleation seeds
- Enhanced mass transfer
- Smaller zeolite crystals





The conversion rate of zeolite A to sodalite increases by a factor of more than 96 compared with hydrothermal synthesis

SEM images



Zeolite A

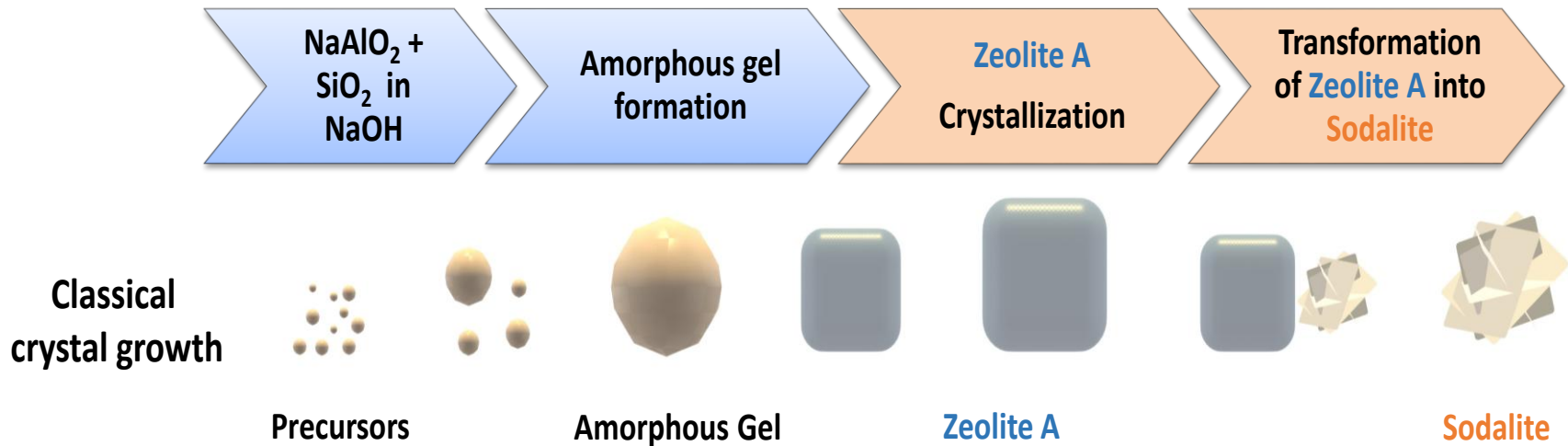
Sodalite
(desert rose)

Conclusion



Fully crystalline zeolite A synthesized under **SHT** conditions in just **25 min** (9.6 times faster than under HT) \Rightarrow **improved crystallization rate**

Smaller zeolite **crystals** with **more homogeneous particle size distribution**



Drastic kinetic increase in the transformation of **zeolite A** into the **more stable sodalite zeolite** (96-fold)

The **combined *in situ* action** of **20 kHz ultrasound** and **hydrothermal conditions** is a **sound alternative** for zeolite synthesis

Acknowledgement



Funding

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PEPR



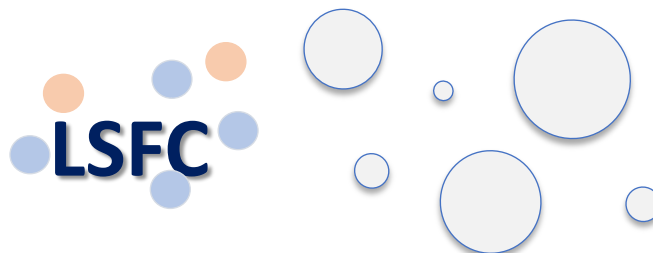
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Supervisors

- Sergey I. Nikitenko
- Tony Chave
- Sabine Valange

SEM images

Joseph Lautru



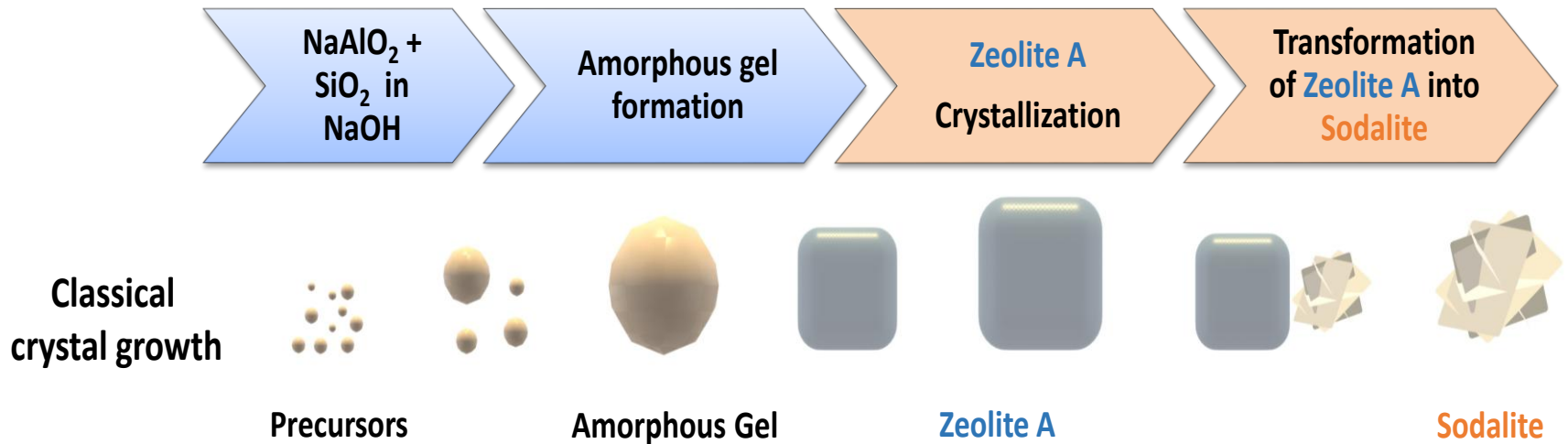
Matthieu
Simon
Rachel
Rita, Julien

Conclusion



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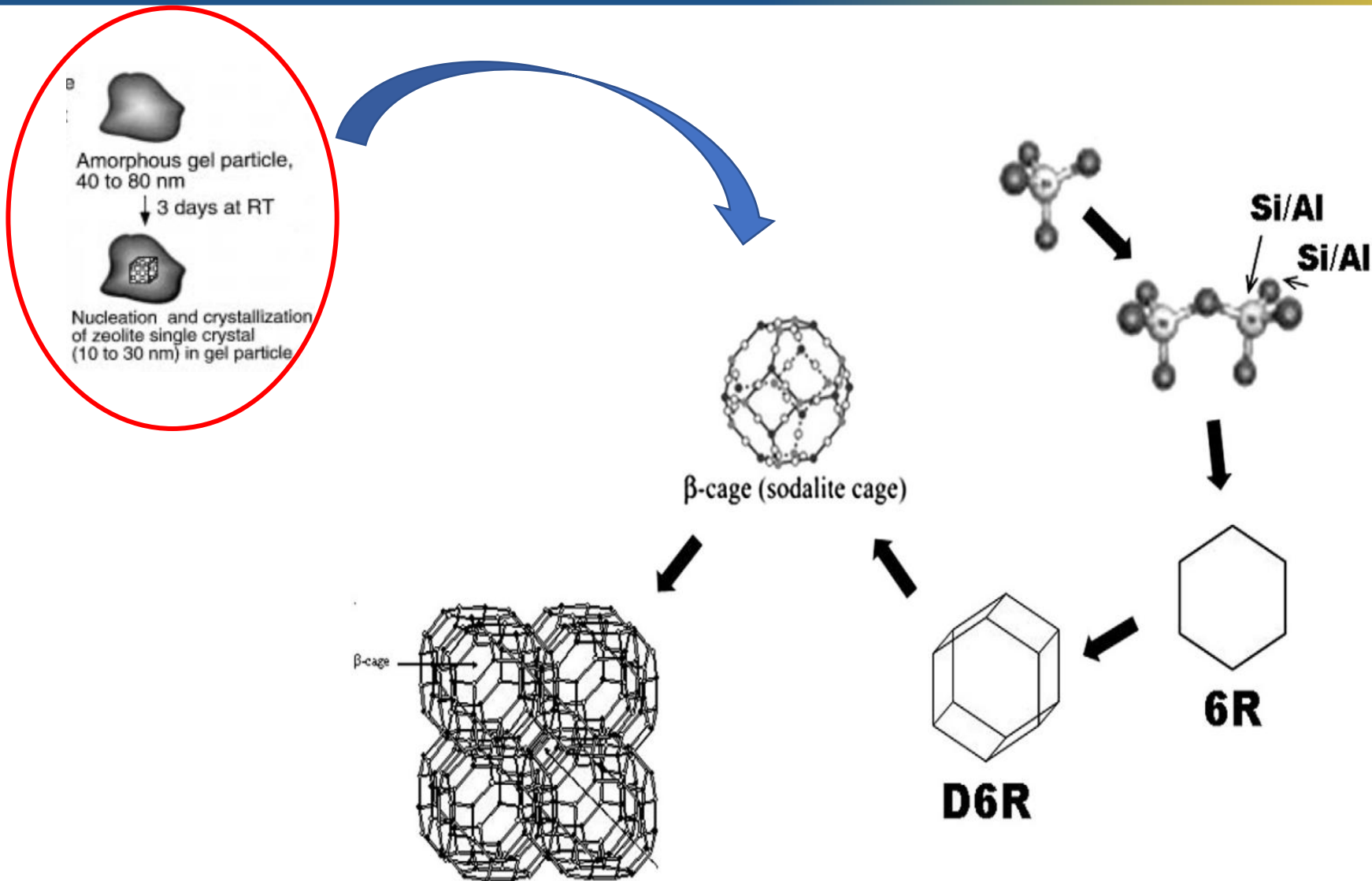
Smaller zeolite **crystals** with **more homogeneous particle size distribution**



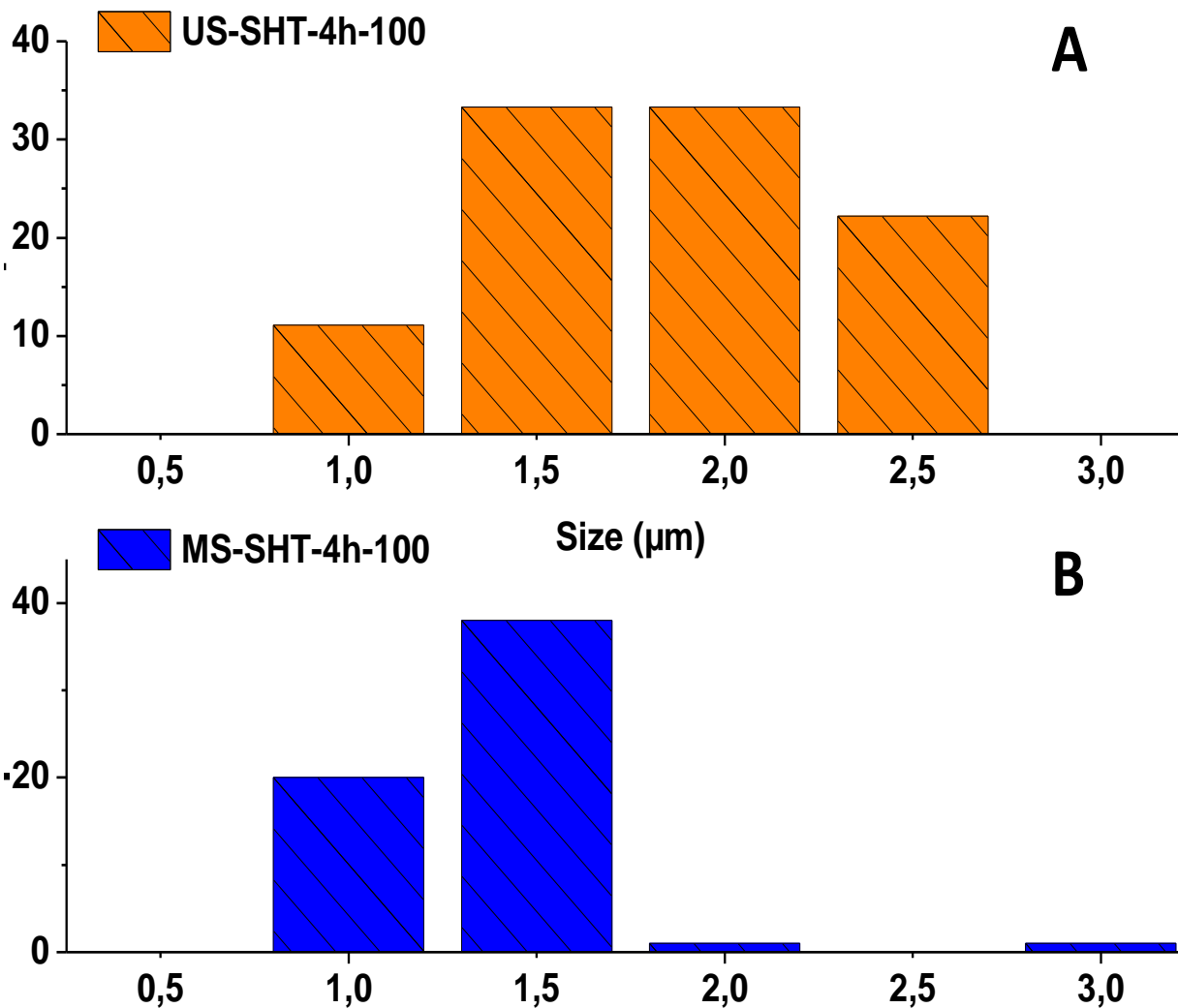
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Classical crystallization of zeolites



Effect on particle size

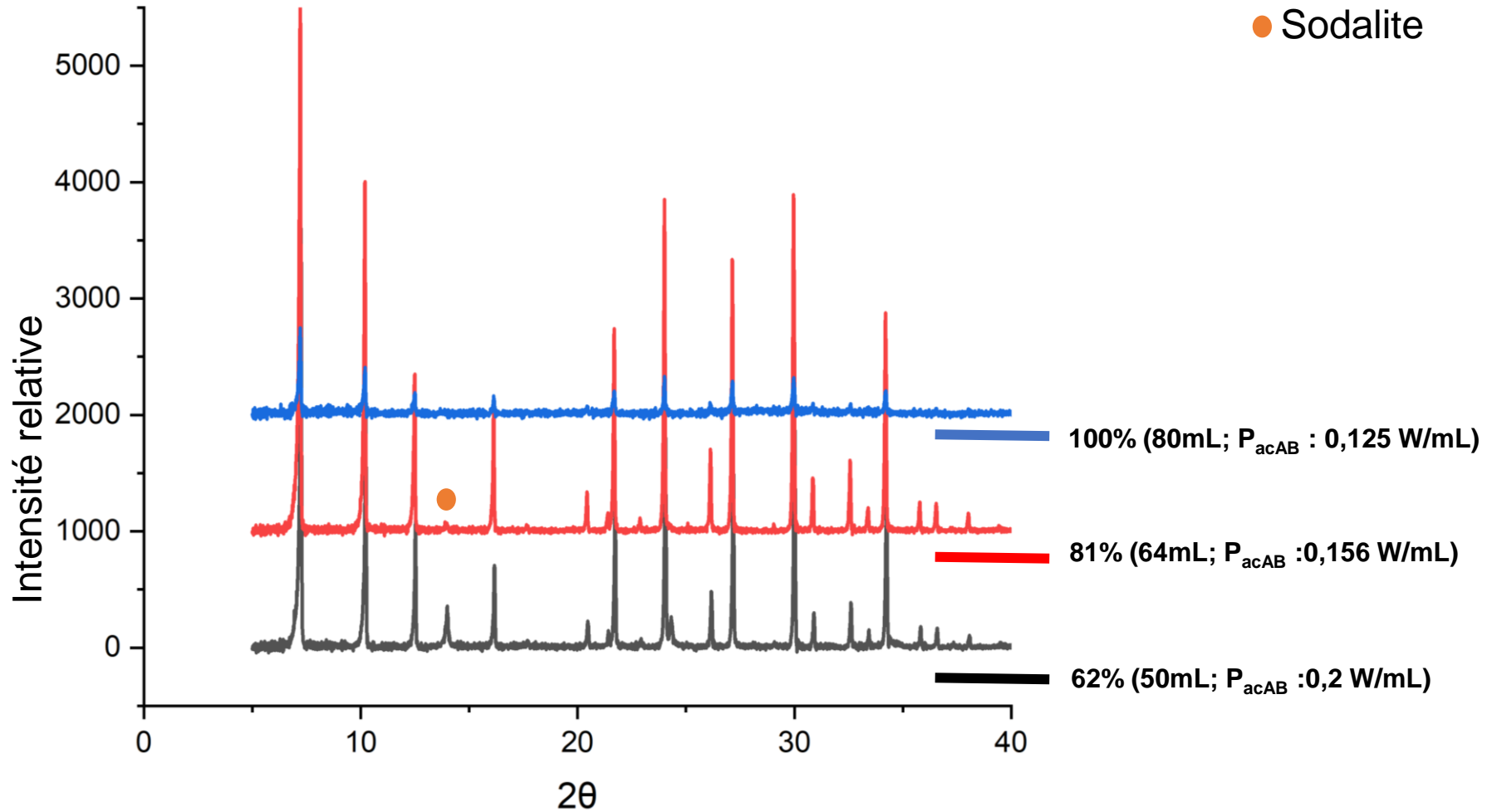


The abundance of sodalite makes it difficult to determine size distribution

Absorbed acoustic power influence



SHT 100 °C_1H
Variation réactor filling rate



Decrease in the conversion rate of amorphous gel to LTA and of LTA to sodalite when absorbed acoustic power decrease