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Influence of gelation kinetics by microwaves

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Motivation

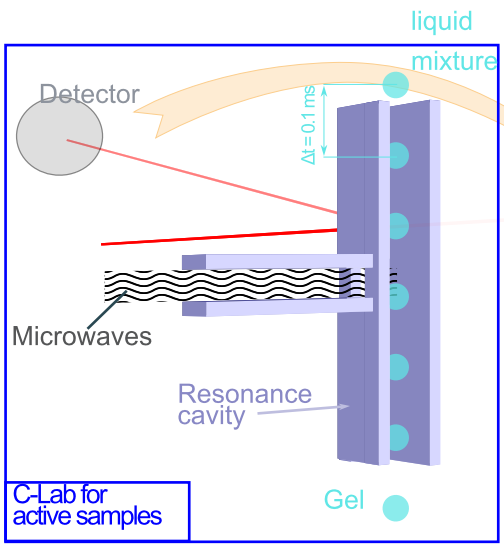
In the PINE project, nuclear fuel microspheres are obtained by microwave internal gelation [1-2]. Free falling droplets, containing chemical ingredients (HMTA, urea and metal nitrate), undergo a precipitation (gelation) induced by microwave heating. The reaction is triggered by decomposing a reactant at a given temperature. However, literature suggests that microwaves could also have a nonthermal catalytic effect on chemical reactions [3-5]. In order to investigate this phenomenon, a X-ray absorption fine structure (XAFS) set-up will be developped where the advancement of the gelation will be monitored in function of the microwave (MW) exposure time.

Sample in a microwave field : agitation of the molecules = heat (+ mass transport?)

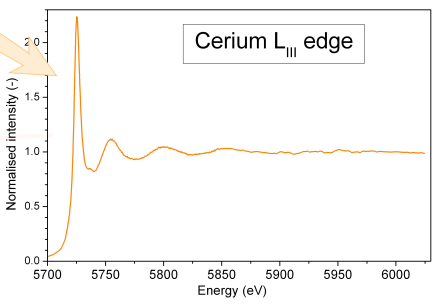
Preliminary Study

At the SuperXAS beamline (SLS, PSI), extended X-ray absorption fine structure (EXAFS) measurements have been performed on trains of droplets.
Since the X-ray absorption spectra are recorded during the reaction, the drop generation and gelation device need to be set up in the hutch. The main aim of these experiments is therefore to verify the applicability of the set up in the SuperXAS beamline, namely to see if a falling microsphere can be detected and analysed.

Basic scheme



EXAFS spectra



...However, the measured signal is obtained for a high number of falling droplets with changing characteristics (size, gelation advancement, ...)

Interaction MW-chemicals

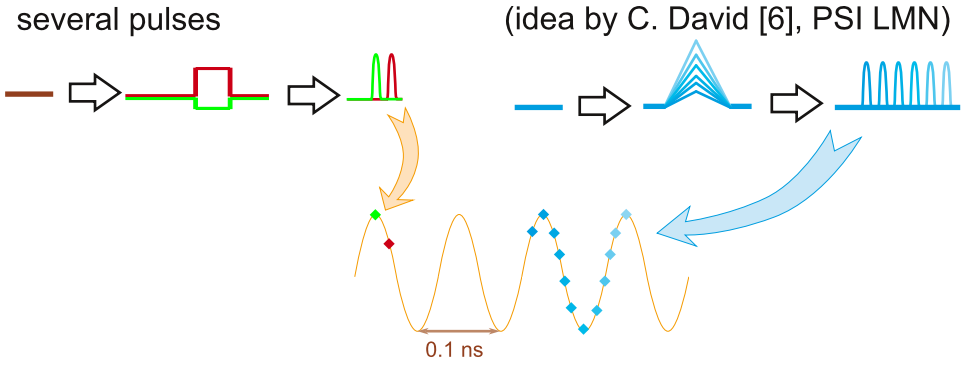
The high time resolution (pulse length 5-20 fs) and peak brillance of the SwissFEL make it possible to perform an X-ray absorption spectroscopy measurements on a single falling droplet (single shot), increasing the quality of the spectra. In the same time, it also allows the determination of effects faster than thermalisation (thermalisation time ~1 ps).
The set-up of the experiment will be similar to that of the SuperXAS one. The experiment can also in a first approach be carried out with an inactive metal surrogate.

Beam Splitter

can be used to produce several pulses

Delay lines

based on diffractive X-ray optics (idea by C. David [6], PSI LMN)



Single XAFS measurements performed on a single period of a microwave (10 GHz) can be produced in the time range of a microwave period (100 ps at 10 GHz). This tool provides a temporal mapping of the surrounding species and resolves the influence of the electro-magnetic field on their interaction.

Parameter requirements

Parameter		Unit	Requirement	Motivation / Remarks
Beam parameters				
Energy		keV	5 to 19	E_{photon} > 17 keV: L3 edge of actinides (U, Pu,...)
Bandw idth		%	~ 1 - 2	a larger bandw idth (4-8%) w ould be beneficial in case of combined spectroscopic and scattering measurements
Beam position	stability	um	< 1	
Beam size		um	< 50	
Photons per initial pulse		#ph	~ 10^{11}	
Pulse length		fs	< 50 fs	To avoid thermalisation effects (ca. 1ps) in the measurement
Beam geometry				
Working distance		mm	> 100	After exit of beam, due to equipment dimensions
Other				
C-lab			yes	In order to perform measurement on active droplets
Synchronization			yes	With a droplet detection device
Delay line			yes	To equally distribute (in time and energy) a single pulse into multiple pulses

Conclusion

The measurements carried out at the SwissFEL will provide us with essential information on the nonthermal microwave contribution with respect to the enhancement of chemical reactions.

Acknowledgements

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