# Elucidation of the $CO_2$ induced Gelation Mechanism of Biopolymer Solutions: Development of In-situ Methods

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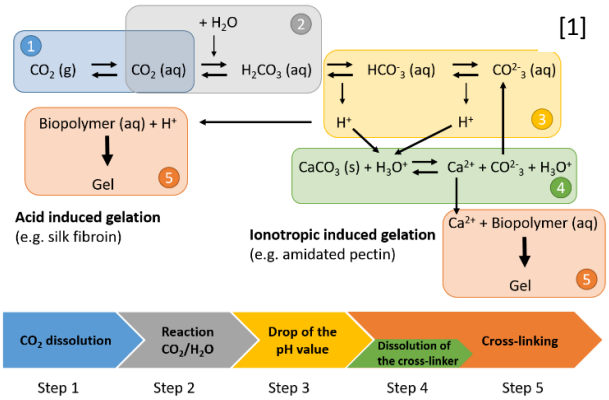
The $CO_2$ induced gelation is a promising gelation method for biopolymer solutions towards the production of biopolymer-based aerogels. It has been shown that the gelation induction by application of pressurized $CO_2$ to divers biopolymer solutions from, e.g. alginate, pectin, silk protein (with or without addition of gelation agents) results in the formation of homogenous and stable hydrogels which afterwards can be processed into aerogels by supercritical drying with $CO_2$.
In this work the overall gelation process of the $CO_2$ induced gelation is divided into five formal steps [1]:




[1]

1) Dissolution of $CO_2$ into the aqueous biopolymer solution
2) Reaction of $CO_2$ and water resulting in formation of carbonic acid
3) Dissociation of carbonic acid and drop of the pH value
4) pH induced dissolution of added gelation agents
5) Gelation induction either directly by pH drop (step 3, without additional gelation agents) or by liberated gelation agents (step 4)

Throughout this work, in-situ measurement methods were developed to investigate the overall gelation mechanism as well as the single steps of the process:

- **In-situ visual gelation observation** to detect turbidity changes and therefore dissolution of gelation agent particles (step 1-4)
- **In-situ pH measurement** for temporal and spatial resolution of pH development via fluorescence measurement (step 1-3)
- **In-situ measurement of $CO_2$ dissolution** for evaluation of uptake equilibrium and kinetics via magnetic suspension balance (step 1)
- **In-situ and ex-situ viscosity measurements** of solution/gel to evaluate the total change of viscosity and gelation kinetics (in-situ: step 5/ ex-situ: step 1-5)

The evaluation of the developed methods was performed with the aqueous gelation system of amidated pectin with $CaCO_3$ added as an additional gelation agent. Via in-situ gelation observation and pH measurement it could be shown that mainly one-dimensional diffusion of $CO_2$ and its reaction products with water occurs inside the gelation sample, resulting in gel formation and drop of the pH value from the contact area with $CO_2$ into the liquid sample. This heterogeneity of gel formation indicates that gel formation itself is much faster than diffusion inside the sample. Further, the impact of the additional carbonate source ($CaCO_3$) directly effects the reaction system of $CO_2$ and water and has a direct impact on the pH development and the overall gelation process and its kinetics [1]. The knowledge obtained opens up opportunities for rational design of homogeneous hydrogels and aerogels using pressurized $CO_2$ as a gelation trigger.

[1] I. Preibisch, L. Ränger, P. Gurikov, I. Smirnova, In Situ Measurement Methods for the CO 2 -Induced Gelation of Biopolymer Systems, Gels. 6 (2020).