

Water-borne poly(meth)acrylates obtained from functional and renewable monomers

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Impact

With the European green deal¹ and the Paris climate agreement², there is an increasing pressure in the reduction of greenhouse gas emissions and transition to a circular economy. This, in combination with other factors such as the slow depletion of fossil fuels, and global unrest resulting in strong fossil energy price fluctuations, the transition away from fossil resources is becoming a subject of discussion. More specifically, in the production of acrylic coatings, the industry relies heavily on the use of fossil derived monomers. Therefore, the exploration of novel biobased monomers is of interest in order to safeguard the production of plastic materials that are of importance in the modern world. By widening the scope of available monomer structures, the transition from fossil to biobased monomers while maintaining tailored polymer properties could be facilitated. Besides renewability, solvent emissions to the environment and waste production related to the production and use of acrylic coatings is undesirable. Therefore, attention has to be paid to polluting factors that are associated with the use of these materials. This can be done by transitioning towards safer and more environmental friendly water-borne systems instead of solvent-based systems, and by improving the recyclability of the final products.

The main objective of this thesis was the development of water-borne acrylic latexes based on biobased and functional monomers for coating and film applications. The polymerization of novel monomer structures can be challenging, especially when the monomers bring added functionality in the form of functional groups. Functional groups can be involved in all kinds of side reactions resulting either in loss of functionality or premature cross-linking of the polymers. Therefore, the polymerization of such novel and functional monomers was systematically investigated and therefore understood. The polymerization conditions that were investigated in a solution type polymerization, were successfully transferred to a water-borne procedure. The resulting polymers retained their functionality to serve as anchor points for further cross-linking reactions.

The functionality of the monomers served additional purpose within the acrylic water-borne polymer. Firstly, cross-linking of the functional polymer latexes improves the physical and thermal properties of the final materials. Secondly, there is a contribution within the scope of reduction in environmental pollution and the circular economy. Using a UV sensitive monomer derived from sugars, we have developed a UV cross-linkable polymer latex, without the use of any Volatile Organic Compounds (VOC's). VOC's are linked to environmental and health related issues.³

Conventional thermosets are cross-linked polymers that cannot be recycled and are disposed after use. Therefore, we have developed a biobased water-borne polyacrylate based on a monomer derived from lignin that is cross-linked using dynamic covalent bonds. The covalent bonds that are formed from an aldehyde and an amine are called imines, which can exchange at elevated temperatures. This results in a material that behaves similar to a conventional thermoset at ambient temperature, but can show macroscopic flow at elevated temperature, enabling the recyclability of these materials. The development of vitrimers is

currently receiving a lot of attention in academia, and is finding ways into industry as well.⁴
⁵ To date, not many vitrimers that are based on biobased and water-borne systems have been investigated. Therefore, this research promotes the application of innovative technologies like vitrimers into a wider application scope consisting of circular products from water-borne polyacrylates.

In this thesis, we have briefly investigated the addition of cellulose nanocrystals (CNC) to the UV curable latex to prepare composite films. The result obtained, indicates towards an affinity between the CNC and latex possibly due to the presence of hydrogen bonding interactions. The composites lead to films consisting of a layered structure at high CNC fractions. Because of this, a drastic increase in mechanical properties of the film are obtained, while only a limited increase in sensitivity to water is achieved. The results show promise for further research towards the barrier performance of such layered composite materials. For example, films and coatings for food packaging and electronics require excellent gas and moisture barrier properties. The use of cellulose in such films is promising due to its properties and abundance of this renewable material. However, the moisture sensitivity is one of the main drawbacks. This research proposes a promising and straightforward solution for the reduction in moisture sensitivity of CNC based films. Despite cellulose, other fillers capable of hydrogen bonding such as chitosan and nanoclays can be investigated in the future for composites with improved physical and barrier properties.

One application of the UV curable latex was investigated as a proof of concept for paper barrier films. The UV cross-linked films show excellent reduction in absorption of oil and water of the paper substrate after exposure compared to uncoated paper. This result highlights a potential industrial application as barrier for paper and cardboard food packaging materials. Nonetheless, the long-term stability, safety, and biocompatibility of the polymers and materials need to be evaluated when food contact or biomedical applications are targeted.

In general, this thesis contributes to the trend of the development of renewable and water-borne systems in the coating industry. Manufacturers of latexes and coating formulations might use the renewable character of their products to promote it to the public with an increasing awareness of circularity. Otherwise, increasing legislative restrictions, or incentives could promote the reduction of fossil materials in the future. The research herein is also published in peer-reviewed scientific journals, which increases its exposure to the industry and scientific community.

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