



Evaluation of natural rubber latex as film forming additive in cementitious mortar

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HIGHLIGHTS

- Addition of 2 wt% natural rubber latex (NRL) dispersion to mortar.
- NRL produces slightly higher compressive and tensile strengths than synthetic EVA.
- Deproteinized NRL shows same effect on mortar strengths as untreated NRL.
- SEM images show NRL intertwines cement hydrates and bridges pore space like EVA.
- Natural rubber latex can be an alternative film forming agent to industrial latex.

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ABSTRACT

The utilization of natural rubber latex (NRL), untreated and treated, to improve the tensile strength of cementitious mortar was studied.

Two NRL samples, a commercial NRL and a laboratory-treated, deproteinized NRL sample, were compared with a commercial synthetic ethylene–vinylacetate (EVA) copolymer with respect to their particle size distribution, minimum film forming temperature (MFFT) and surface charge.

Furthermore, cement mortars containing 2 wt% of the latexes were tested for their compressive and tensile strength development after 3–14 days. Both NRL samples increased tensile strength as much as the synthetic EVA latex. Thus, deproteinisation of NRL is not required.

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1. Introduction

Latex dispersions represent an important group of chemical admixtures used in cement based building materials. They are applied as organic binders in paints and coatings and are also suitable as co-binders in combination with mineral binders, e.g. cement. There, typical applications of latex polymers include tile adhesives and grouts, adhesive and repair mortars, waterproofing membranes or self-leveling underlayments (SLUs) [1]. Due to their ability to form flexible and homogeneous polymer films, latex polymers provide cohesion of the fresh mortar and adhesion on various substrates. Furthermore, they increase the flexural strength of the hardened mortar [2,3]. So far, the latex dispersions applied in the construction industry are exclusively synthetic ones, most of them are based on ethylene/vinylacetate, styrene/butadi-

ene or acrylate chemistry and are produced via emulsion polymerization [4].

Similar to synthetic polymer dispersions, natural rubber latex (NRL) also has the ability to form homogeneous and flexible films upon dehydration.

Since the 1880s natural rubber latex constitutes a widely used raw material which is applied in many aspects of daily life [5].

Freshly tapped natural rubber latex consists mainly of two components, the polymer poly(*cis*-1,4-isoprene) and water. The concentration of the polymer varies between 25 and 40 wt%, the remainder being essentially of water. Additionally, some minor components such as proteins, lipids, carbohydrates, organic solutes and inorganic substances are contained in the serum [6,7]. The protein concentration in the latex dispersion lies in the range of 1–2 wt%, 20–30% of them are adsorbed on the rubber particles [7]. NRL consists of spherical latex particles which are surrounded by a mixed layer of adsorbed proteins and phospholipids [8,9]. These components play an important role in stabilizing the latex parti-

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cles. NRL is commonly treated with ammonia, thus the interfacial proteins and lipids become negatively charged. This way colloidal stability of the latex particles is achieved [7]. Furthermore, ammonia prevents the proliferation of bacteria in NRL which attack the protein layer of the rubber latex particles and result in destabilisation of the latex [10]. For transportation and processing purposes, the freshly tapped NRL normally is concentrated up to about 60 wt% solids content.

Proteins may retard the hydration of cement. To eliminate this undesired effect, the proteins can be removed by deproteinisation of the NRL with the help of surfactants such as, for example sodium dodecyl sulfate (SDS). The proteins adsorb the surfactant and massive cooperative binding is observed which leads to protein denaturation. This denaturation disrupts the non-covalent, structure-stabilizing bonds of the proteins. As a result, their secondary and tertiary structure are destroyed [11]. The denaturated proteins become water-soluble and are released into the solution. The surfaces of the NRL particles are then covered by SDS molecules instead of proteins which stabilize the particles and prevent particle coagulation.

NRL represents an eco-friendly, sustainable and non-petroleum based material. Low cost, increasing availability and the ecological aspect as a renewable material make natural rubber attractive for new applications [5]. In spite of its advantages, successful use of natural rubber in cementitious mixtures has not occurred so far.

Only limited previous work exists on the use of NRL in cementitious mixtures such as concrete or mortar.

For example, Bala et al. studied the influence on the compressive strength from different substances contained in natural rubber latex [12]. Interestingly, they found that small differences in the dry rubber content does not play an important role for improvement in the compressive strength. Also, the proteins in the NRL do not affect the compressive strength, but the volatile fatty acids and metals, particularly zinc, reduce the strength. Overall, the compressive strength of concrete was improved only marginally by the addition of NRL. The authors noticed as well that the compressive strength of natural rubber modified concrete decreases with increasing temperature [13]. They also found that addition of NRL into concrete transforms the porous microstructure of conventional concrete into a denser matrix [14]. Yet, the effect of natural rubber latex on the tensile strength was not been tested there.

In this work, the general applicability of NRL in a standard cement based mortar was evaluated. Different latex dispersions were characterized, namely a commercial concentrated NRL sample, a deproteinized NRL sample and a synthetic industrial EVA sample. Their solids contents, particle size distributions and pH values were determined. Colloidal stability was assessed via zeta potential and their minimum film forming temperatures (MFFT) were determined using a Kofler bench. Furthermore, performance of the three dispersions in a basic cementitious mortar formulation and a reference mortar without latex was assessed with respect to compressive and tensile strengths at comparable consistency and air content. Finally, the latex films formed in the hardened cement mortars were visualized by scanning electron microscopy (SEM).

2. Materials and methods

2.1. Materials

2.1.1. Latex dispersions

The natural rubber latex sample (solids content 61.5 wt%) was a commercial, low ammonia treated latex supplied by the Rubber Authority of Thailand, Phatholyothin rd. Chatuchak, 10900 Bangkok, Thailand. A commercial EVA latex sample (solids content 51 wt%) was provided by Dow Olefinverbund GmbH, Schkopau, Germany.

2.1.2. Sodium dodecyl sulfate

Sodium dodecyl sulfate (SDS) was received from Merck KGaA, Darmstadt, Germany. SDS was applied for the deproteinisation of NRL and was also added to the mortar to introduce air bubbles.

2.1.3. Cement

The cement sample was an ordinary Portland cement CEM I 52.5 R from Schwenk Zement KG, Germany which is popular used as base cement. Its average particle size (d_{50} value) was obtained by laser granulometry (Cilas 1064, Cilas Company, Marseille, France) and was found at 5.92 μm . The specific density of the cement was 3.18 kg/L as measured via Helium pycnometry (Ultrapycnometer 1000, Quantachrome Instruments, Boynton Beach, USA). Table 1 shows the phase composition of the cement sample as determined by quantitative X-ray diffraction (Bruker AXS D8 Advance, Karlsruhe, Germany) using Rietveld refinement (software Topas 4.0).

2.1.4. Defoamer

When using polymer dispersions in mortar, addition of a defoamer is necessary because the latex introduces air. In this work, three different defoamers were used. Tributyl phosphate was supplied by Fluka Chemika while Surfynol[®] MD-20 was obtained from Air Products, Netherlands and Dowfax[®] DF 141 was furnished by Dow Chemical Company, Midland, Michigan, United States.

2.1.5. Superplasticizer

For the reference mortar and the mortar formulations containing the NRL samples a superplasticizer had to be used. For this purpose, a self-synthesized, methacrylate ester based (MPEG type) polycarboxylate from our laboratories was applied. Its synthesis is described in Ref. [15]. It was composed of methacrylic acid and MPEG methacrylate ester at a molar ratio of 6:1 and contained a side chain made of 45 ethylene oxide units.

2.2. Methods

2.2.1. Deproteinisation of NRL

Commercial NRL with a solids content of 61.5% was diluted with DI water to 30 wt% and deproteinized by allowing it to react with 1 wt% sodium dodecyl sulfate (SDS) for 10 min in an ultrasonic bath, followed by centrifugation at 8500 rpm [16,17].

The upper rubber fraction (30 vol%) was collected and re-dispersed in 1 wt% SDS solution to make 30 wt% dry rubber content and then was again treated in an ultrasonic bath for 10 min and centrifuged at 8500 rpm. This procedure was repeated four times. The final upper fraction was collected and dispersed in distilled water. The solids content of this NRL-SDS sample was 42.4 wt%.

2.2.2. Characterization of the dispersions

2.2.2.1. Solids content of liquid latex. The solids content of the latex dispersions was determined on a MA-30 infrared drying balance from Sartorius, Hamburg, Germany.

2.2.2.2. pH value. The pH value was determined on a pH 11 instrument from Schott, Mainz, Germany.

2.2.2.3. Particle size distribution. Particle size distribution of the dispersions was determined via dynamic light scattering (DLS) utilizing a Zetasizer Nano ZS apparatus from Malvern Instruments, Worcestershire, UK.

Table 1

Phase composition of the CEM I 52.5 R sample, as determined via Q-XRD using Rietveld refinement.

Phase	Content [wt%]
C ₃ S	53.62
C ₂ S	17.49
C ₃ A, cubic	7.48
C ₃ A, orthorhombic	1.74
C ₄ AF, orthorhombic	8.03
CaSO ₄	3.64
CaSO ₄ ·0.5 H ₂ O	2.14
CaSO ₄ ·2 H ₂ O	0.00
Calcite	3.46
Quartz	0.77
Dolomite	1.63
Total	100.00

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