

CHAPTER I

INTRODUCTION

1.1 Statement of Problems

In rubber industry, besides carbon black, silica is the other reinforcing filler used to impart specific properties to rubber compounds. Silica or $(\text{SiO}_2)_n$ offers a number of advantages on the vulcanizates such as good tear strength, good abrasion resistance and reduction in heat build-up [1]. However, mechanical mixing between silica and rubber is somewhat a challenge due to the polar hydroxyl groups on the silica surface. Its presence can result in strong silica-silica interaction by hydrogen bonding. This can lead to aggregation of silica particles inside the rubber matrix [2], often resulting in a poor dispersion of these particles and high viscosity during mixing. The silica can also adsorb curing agents and accelerators on its surface which results in the reduction of curing efficiency [3]. The other important difficulty is the incompatibility of inorganic silica with the organic rubber.

These problems are partially overcome or minimized by specific additives and mixing procedures. As a method for overcoming these difficulties, sol-gel process of tetraethoxysilane (TEOS) has been developed in order to generate silica inside the rubber matrix, instead of mixing them. The sol-gel process was applied to the preparation of organic-inorganic hybrid materials, which synergistically combine the best properties of organic materials with the excellent properties of inorganic glass. Therefore, this process has been extensively applied to a variety of polymers, such as styrene-butadiene rubber (SBR) [4], butadiene rubber (BR) [5], acrylonitrile-butadiene rubber (NBR) [6], epoxidized natural rubber (ENR) [7], polyimide (PI) [8], and polyacrylate [9].

Alkyltrialkoxysilanes are used to chemically modify silica surfaces by sol-gel process, in order to improve the filler dispersion and prevent adsorption of curing agents on the silica surface. The alkyl groups on silica particle surfaces can prevent

the aggregation of the particles due to their steric repulsion. Some functional groups such as alkyl, amine, epoxide, and vinyl groups can be introduced by selecting the right choice of alkyltriethoxysilane [8, 10].

Until recently, the sol-gel reaction of TEOS was carried out in rubber latex for compounding rubber and silica. The composite containing *in situ* silica had a higher tensile modulus and tear strength than the composite prepared by conventionally mixing with silica powder [11]. When adding bis-(3-triethoxysilylpropyl)tetrasulfide (TESPT) together with TEOS into the latex, mechanical property of NR-silica vulcanizate was improved [12].

In this work, the focus was to generate modified silica particles inside rubber matrix. TEOS were used in combination with three types of alkyltriethoxysilanes to prepare the *in situ*-silica filled NR vulcanizates. It was hypothesized that the *in situ* generated silica particle having alkyl group can improve the dispersibility and mechanical properties of silica-NR vulcanizate.

1.2 Objective

This work introduces alkyltriethoxysilanes and tetraethoxysilane (TEOS) to generate silica by sol-gel process of the two silanes in NR latex. Four types of ethoxysilanes; tetraethoxysilane (TEOS), vinyltriethoxysilane (VTOS), ethyltriethoxysilane (ETOS), and *i*-butyltriethoxysilane (BTOS), were studied. Silica particle distribution, morphology and mechanical properties of the *in situ* silica-filled NR vulcanizate were analyzed.

1.3 Scope of the Investigation

The stepwise investigation was carried out as follows:

1. Preparation of NR-silica composites by sol-gel process of TEOS and alkyltriethoxysilanes in concentrated natural rubber latex (%dry rubber content = 60).

2. Determination of curing behavior and mechanical properties of the NR-silica vulcanizates according to ASTM methods.
3. Characterization of the NR-silica particles for
 - morphology of the *in situ*-generated silica by SEM and TEM.
 - spectroscopic evidences of modified *in situ*-generated silica by ^{29}Si -CPMAS NMR.



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