CHAPTER I

INTRODUCTION

1.1 Statement of Problems

Natural rubber is an important agricultural commodity essential for the manufacturing. Asia is the center of production and Thailand is the biggest producer today [1]. The world production of natural rubber (NR) has increased at an average rate of 3.74 percent over the five years period (1994-1998). The average production of natural rubber is 6.22 million tons per year, while the world consumption during that period is 6.17 million tons per year [2]. Being the largest producer of natural rubber, the production in Thailand is expected to reach 2.12 million tons in the year of 2001. The global natural rubber production is expected to grow at 1.1 percent during 1999-2001, reaching about 8,535 thousand metric tons in 2010 [3].

Natural rubber is the strongest of all rubbers and has excellent dynamic properties such as resistance to fatigue but it is less resistant to environment damage (*e.g.* by ozone in the atmosphere and oils) than are some synthetic rubbers. In some products the choice of rubber is determined solely by properties (*e.g.* aircraft tyres which require 100 per cent natural rubber) but in many products there is a competition between natural and synthetic rubbers on the basis of price and properties.

Due to the industrial growth, the technology and the quality of the products from natural rubber have been greatly developed for the past decade. Reinforcement of natural rubber by silica filler has been of interest for many applications where colorless or clear rubber is desirable. Adverse mechanical properties of silica-filled natural rubber are often problematic due to poor adhesion between natural rubber matrix and polar silanol groups on silica surface. Several approaches have been used to promote adhesion. One method is to lower interfacial energy at the interfaces, including chemical modification of natural rubber by oxidation or grafting of polar monomers to increase rubber polarity. Another method involved surface treatment of silica filler to improve compatibility by introducing hydrophobic moieties. Pre-coating of silane coupling reagent on silica was also found necessary for uniform surface coating that can yield improved crosslinking density. Another promising way to improve adhesion is to generate interfacial chemical bonding by adding silane coupling reagents during the curing process.

Therefore, the main objective of this research is to introduce an alternative approach to forming crosslinked network at the "interphase" between natural rubber and silica. The research studies can be divided into two parts. The model study involved chemical modification of silicon oxide (a flat surface model for silica) by vinyl-containing silane reagents followed by reaction with squalene (a natural rubber model) under heat in the presence of an appropriate catalysts. Comparative studies were also carried out using a saturated analog of squalene, squalane. Chemical composition and thickness of interfacial crosslinked network can be conveniently assessed by x-ray photoelectron spectroscopy (XPS) and ellipsometry, respectively. This model study should provide insightful information for understanding the reactions at the "interphase" between modified silica and natural rubber, which eventually affect curing. The second part of research involved the determination of curing behavior and mechanical properties of natural rubber composites filled with vinyl-containing silica using sulphur as a curing agent. This part of research was carried out in order to consider the feasibility of using the surface-modified silica for practical uses.

1.2 Objectives

1. To study the effects of silane reagent and catalyst on the extent of interfacial crosslinking in terms of thickness of the crosslinked network at the interface between squalene and vinyl-containing silicon oxide surface.

2. To determine the curing behavior and mechanical properties of natural rubber composites filled with vinyl-containing silica.

1.3 Scope of the Investigation

The stepwise investigation was carried out as follows:

1. Literature survey for related research work.
2. To prepare vinyl-containing silicon oxide by chemical modification of silicon oxide surface with vinyl-containing silane reagents and to characterize the modified substrate by contact angle measurement and x-ray photoelectron spectroscopy (XPS).
3. To react vinyl-containing silica by reactions with squalene or squalane under heat in the presence of a number of catalysts.

4. To study the effect of silane reagents and catalysts on the thickness of the crosslinked network at the interface between squalene and vinyl-containing silicon oxide surface and to analyze the resulting surfaces by ellipsometry and x-ray photoelectron spectroscopy (XPS).
5. To determine the curing behavior and mechanical properties of natural rubber composites filled with surface modified silica according to ASTM methods.
6. To summarize the results.