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Preparation of Composites of Citrus Flavonoids and Various Porous Silica Materials and their Performance as Ultraviolet Absorbers for Sunscreens

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Abstract

Flavonoids from the citrus fruit shekwasha were adsorbed on two types of porous silica materials, mesoporous silica and amorphous silica gel, in order to improve the ultraviolet A (UVA) absorption capacity and prevent the penetration of flavonoids into the skin. Mesoporous silica showed a higher adsorptive capacity of flavonoids than the amorphous silica gel, owing to its high surface area and uniform mesoporous structure. Both the composites (in powdered forms) exhibited UVA absorption, but the silica gel composite also showed absorption in the visible range, making it less suitable as a UVA absorber. The oil dispersions of the mesoporous silica composites in the form of thin films also absorbed UVA effectively, indicating that the mesoporous silica composite with the shekwasha flavonoids can be used as a new UVA absorber in skin cosmetics.

Key-words: *Citrus depressa, Flavonoids, Silica gel, MCM-41 mesoporous silica, UV-vis absorbance*

1. Introduction

Ultraviolet (UV) radiation can be classified on the basis of wavelength as UVA (320-400 nm), UVB (280-320 nm) and UVC (100-280 nm). Among these, UVA is responsible for causing skin tanning and premature skin aging such as wrinkles and dark spots. Hence, various UVA protective agents are used in daily use cosmetics such as lotions and creams.

Ultraviolet protective agents (sunscreen materials) for cosmetics can be organic or inorganic. Inorganic materials like titanium oxide and zinc oxide are known to be generally stable and cause less skin problems. However, they are not transparent (too white) and tend to cause a rough, 'powdery' feeling when applied on the skin, making them less desirable for use as UVA protection cosmetics.

Organic materials such as avobenzone, octylcrylene, and octinoxate are transparent and oil soluble, but are likely to cause skin irritation as they can penetrate the skin[1].

Flavonoids are natural organic compounds present in many kinds of fruits. The flavonoids derived from the citrus fruit shekwasha (*Citrus depressa*) contain polymethoxylated flavones (PMFs), mainly nobiletin and tangeretin (**Fig. 1**), which are recognized for their antioxidant and anti-inflammatory properties. In addition, their solutions effectively absorb UVA and are also transparent (colorless), thereby making them attractive natural UVA protection agents. However, reports indicate that they can cause skin irritation[2].

Porous silica materials such as mesoporous silica (MPS), zeolites, and amorphous silica gel are widely used to stabilize organic molecules by adsorbing them into their pores. Among these, MPS composites are well-known due to their high surface area (>1000 m^2/g), and well-ordered and tunable uniform pore sizes (1-30 nm)[3] which are appropriate for fitting the molecular structures of PMFs.

In this study, one such popular mesoporous silica of the type MCM-41 was prepared by the hydrothermal synthesis method and used to adsorb PMFs from shekwasha flavonoids. The obtained PMF-MPS composites (with different amounts of adsorbed PMFs) were then investigated as UVA protective agents, by comparing their adsorption efficiency and UV-vis absorption performance with that of amorphous silica gel based PMF composites. In order to be ideal UVA protective agents for use in cosmetics, the PMFs must first be highly adsorbed and uniformly dispersed within the pores of the MPS. They must also absorb UVA effectively, without absorbing visible light (therefore be colorless) in their composite powder forms. In addition to this, the composites must exhibit effective UVA absorption performance even when dispersed in thin oil-based films, similar to actual application on the skin.

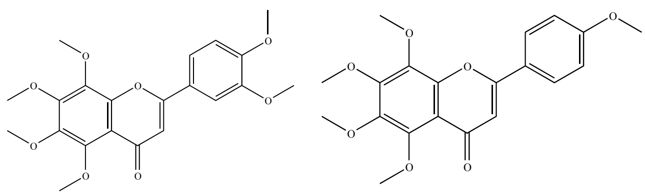

Fig. 1 Chemical structure of PMFs.