

DEVELOPMENT OF IONIC LIQUIDS MEDIATED NOVEL FOOD PRESERVATION SYSTEM BY ENHANCING SOLUBILITY OF FLAVONOIDS

エムディー, シムル, イスラム

<https://hdl.handle.net/2324/6787580>

出版情報 : Kyushu University, 2022, 博士 (工学), 課程博士
バージョン :
権利関係 :



氏 名 : MD. SHIMUL ISLAM (エムディー シムル イスラム)

論文名 : DEVELOPMENT OF IONIC LIQUIDS MEDIATED NOVEL FOOD PRESERVATION
SYSTEM BY ENHANCING SOLUBILITY OF FLAVONOIDS
(イオン液体によるフルボノイド類の溶解性向上を利用した新たな食品保存
システムの開発)

区分 : 甲

論文内容の要旨

Proper food preservation techniques can feed 2 billion hungry people, prevent 40000 deaths from foodborne disease, and reduce more than 30% food loss. Conventional preservation methods including chemical preservatives change organoleptic properties of food, cause nutrient loss and possess some health risks. Recently, natural antimicrobial agents are considered as a potential green alternative to chemical preservatives and other food preservation methods. Many of the plant derived compounds which are known as flavonoids have potential chemical and biological properties including antioxidant and antimicrobial activities. Functional applications of flavonoids are limited due to its low solubility, poor stability, and low bioavailability. Different modern techniques such as nanoemulsion, nanoparticles etc. are used to overcome those limitations. Some of the modern delivery system can't deliver flavonoids at required doses at recommended amount of carrier materials. Ionic liquids (ILs) as highly biocompatible, biodegradable and environmental friendly green solvent could be potential marrier materials to deliver flavonoids in food systems.

In the **second chapter**, we have devised to screen and synthesize bio-based phenolic acids bearing amino acid ester ILs as a potent green solvent for dissolving Luteolin (Lut) for food processing applications. Twenty cations (standard amino acid ethyl esters) and nine anions (phenolic acids) were selected and assessed for dissolution of LUT via COSMO-RS. To address the poor aqueous solubility of flavonoids, we have synthesized and characterized four novel proline ethyl ester-based phenolic acids ILs (PEEP-ILs) i.e., IL[ProEt][Fer], IL[ProEt][Cou], IL[ProEt][Van], and IL[ProEt][Ben]. The PEEP-ILs (60% IL in water) increased aqueous solubility of LUT by 207 to 879 folds compared with that in water, indicating the possibility of utilization of LUT in the aqueous system. ^1H NMR assessment confirmed that the ILs facilitated to solubilize the LUT through multiple hydrogen bonding, π - π , and cation- π interactions between the LUT and the ILs. The biocompatibility study revealed that PEEP-ILs were relatively harmless. COSMO-RS prediction of LUT solubility showed good agreement with experimental solubility in PEEP-ILs. LUT with newly synthesized PEEP-ILs retained better organoleptic properties of red apple piece. These results clearly suggest that PEEP-ILs

can be potential green alternatives to conventional toxic solvents for bioactivity of poorly water-soluble natural preservatives.

Formulation based encapsulation has become a cutting-edge method due to their capacity for self-assembly, controlled release profile, and high encapsulation efficiency. As the Lut is photosensitive molecule, their encapsulation by carrier materials can improve their bioactivity. In the **third chapter**, we have focused on to formulate and investigate nano-sized micellar formulation of Lut using a low carrier material concentration to enhance LUT dissolution and bioactivity. The LUT-loaded micellar formulation (LUT-MF) was prepared using the solid-in-water nanodispersion technique utilizing biocompatible and low toxic surface-active IL choline oleate (1.5%). Dynamic light scattering (DLS) and transmission electron microscopy (TEM) confirmed that the LUT-MF formed spherical micelles with a mean particle size of approximately 73 nm. LUT-MF showed excellent encapsulation efficiency (94.3%) and enhanced the aqueous solubility. An *in vitro* release study in PBS containing 30% ethanol demonstrated 3.94 times higher release of LUT from LUT-MF than from LUT suspension after 24 h. The LUT-MF showed better antioxidant activity than both free LUT and ascorbic acid and higher antibacterial activity than sodium benzoate. Food preservation studies revealed that treating strawberries with LUT-MF effectively preserved color and prevented shriveling and decay over 8 days storage at 10°C, compared with the sodium benzoate treatment. The results suggest that LUT-MF is a potential natural preservative alternative to conventional toxic chemical preservatives used in the food industry.

Synergistic potentialities of flavonoids give plants a stronger defense against different diseases and serve as potent antimicrobials or immunological boosters. To utilize synergistic properties of flavonoids, we concentrated in the **fourth chapter** to develop choline oleate assisted micellar formulations for luteolin, quercetin and naringenin in combinations to increase their solubility and bioactivity. The developed micelles of Lut, Nar, and Quer were encapsulated, in amorphous state and spherical in shape. Nar-MF, LN-MF, and LNQ-MF released 80% of their component within 4 hours and maintained those concentration up to 12 hours. [Cho][Ole] based formulations showed better antioxidant activity than that of free Nar and Quer. Combinations of Lut with Nar (LN-MF), and Lut with Nar and Quer (LNQ-MF) demonstrated synergistic or additive effects on antimicrobial activities and prevented the coagulation of milk from microbial metabolism. ILs derived from natural sources could replace conventional solvents and act as carrier materials for the delivery of poorly water-soluble flavonoids in the food industry. However, we should concentrate more on the optimization of the formulations for better performance and their safety should be checked extensively on animal model.