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Formulations of galangin and its derivatives with antimicrobial, anticancer, and anti-neurodegenerative effects: Recent advances and challenges

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ABSTRACT

Numerous biomedical activities including antioxidant, anti-inflammatory, antimicrobial, antiproliferative, anti-obesity, anti-genotoxic, anti-neurodegenerative, and antidiabetic activities have been indicated for galangin flavonol. As the main application, galangin can be utilized as a potential adjuvant for reducing the cytotoxic effects of chemotherapeutic agents such as cisplatin and doxorubicin. Blocking p38 mitogen-activated protein kinases, nod-like receptor protein 3 signals, and nuclear factor-kappa B, have been found for the main anti-inflammatory mechanisms of galangin. Several disadvantages including sensitivity to light, temperature, pH, low insolubility in aqueous medium, and semi-permeability to gastrointestinal barriers have been indicated for galangin. For overcoming these clinical limitations, presenting efficacious formulations composed of micro or nanomaterials is critical. This mini-review made an effort to discuss the advances and challenges of these formulations with anti-inflammatory, anticancer, anti-neurodegenerative, antibacterial, antiviral, and anti-osteoporosis activities for opening novel avenues of future investigations.

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Introduction

Exploring new natural compounds with promising therapeutic effects might open new perspectives in biomedicine [1, 2]. Galangin (3,5,7-trihydroxyflavone) is a flavonol derivative without hydroxyl group in the B-ring (Figure 1) [3]. This flavonol metabolite can be extracted from some plant species such as *Alpinia officinarum*, *Alpinia galangal*, and *Helichrysum aureonitens* [4, 5]. Abundant biomedical activities including anti-inflammatory, antioxidant, antibacterial, antiviral, antifungal, antiproliferative, anti-obesity, anti-genotoxic, cardiovascular protective, cerebrovascular protective, anti-neurodegenerative, anti-osteoporosis, and antidiabetic activities have been indicated for this metabolite [4, 6-9]. Several clinical limitations including sensitivity to light, temperature, pH, and low insolubility in aqueous medium and semi-permeability to gastrointestinal barriers have been reported for galangin [10]. Modern nanotechnology using different nanomaterials in nanomedicine may overcome these disadvantageous characteristics of galangin [11].

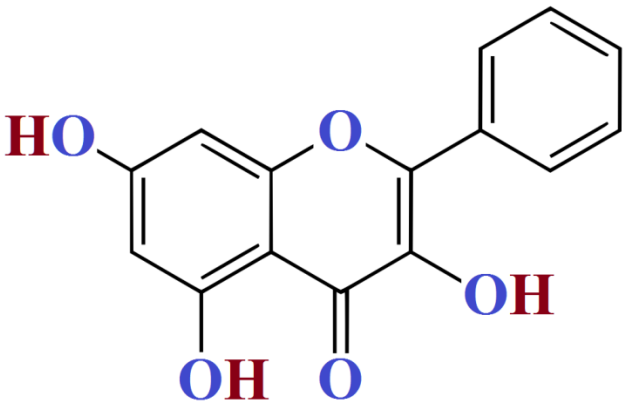

Fig. 1. Chemical structure of galangin.

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Anti-inflammatory effect

Hindering p38 mitogen-activated protein kinases (MAPK), nod-like receptor protein 3 signals, and nuclear factor-kappa B (NF-κB), have been found for the main anti-inflammatory mechanisms of galangin [12]. A potential way to therapy neurodegenerative diseases is by controlling microglia-associated neuroinflammation [13]. The expression of pro-inflammatory cytokines and inducible nitric oxide synthase (iNOS) can be hindered by galangin. This flavonol can increase the expression level of anti-inflammatory interleukin (IL)-10 in lipopolysaccharide (LPS)-stimulated BV2 microglia. In addition, c-Jun N-terminal kinase (JNK), NFκB, LPS-induced phosphorylation of p38 MAPK, and phosphatidylinositol 3-kinase (PI3K)/protein kinase B (Akt) activity may be decreased by galangin. Furthermore, galangin enhanced the activity of transcription factors including cAMP response element-binding protein (CREB), peroxisome proliferator-activated receptor--γ (PPAR-γ), and nuclear factor-E2-related factor 2 (Nrf2) have been found as anti-inflammatory mechanisms of galangin [14].

Anti-neurodegenerative effect

In neurodegenerative disorders such as Alzheimer's disease and Parkinson's disease, neuroinflammation has a fundamental effect characterized by the enhanced expression of inflammatory proteins such as matrix metalloproteinases (MMPs) [15]. LPS can augment MMP-9 expression via the activation of numerous transcription factors such as forkhead box protein O1 (FoxO1) and activator protein 1 (AP-1). Interestingly, galangin can decrease neuroinflammation by suppressing LPS-induced MMP-9 expression in rat brain astrocytes [16]. In the case of Parkinson's disease, *in silico* study showed that galangin had a low binding energy of -9.4 kcal/mol towards the active site of α-synuclein [17]. In neurodegenerative disease such as Parkinson's diseases, neuroinflammation results from microglial activation. Therefore, hindering pro-inflammatory cytokines and microglial activation is critical to the therapy of neurodegenerative diseases. In this regard, galangin reduced the production of nitric oxide, inducible nitric oxide synthase, and IL-1β in LPS-stimulated BV-2 microglial cells. This metabolite showed an anti-neuroinflammatory effect on LPS-stimulated BV-2 microglial cells through the NF-κB and MAPK signaling pathways [18].

Anticancer effect

The cell proliferation rate in the HCT-116 cell line (a human colorectal carcinoma cell line) has been reduced upon treatment by galangin in a time-dependent way via blocking effect on HCT-116 clonogenicity and inducing cell cycle arrest at the G2/M or G1 phase [19]. Galangin can suppress cell cycle progression, MAPK, Akt, or mammalian target of rapamycin (mTOR) activity resulting in apoptotic cell death via the production of caspase-9/8/3. This metabolite can block tumor invasion and metastasis via reducing the upregulation of matrix metalloproteinase-2/-9 (MMP-2/-9) [20]. In a dose-dependent way, galangin inhibited the expression of CD44 and blocked the angiogenesis of glioma cells via downregulating vascular endothelial growth factor (VEGF) in human umbilical vein endothelial cells (HUVECs) [21]. Numerous synthetic and organic nanomaterials have been indicated as promising nanocarriers for the encapsulation, loading, and delivery of drugs or natural therapeutic agents [22, 23]. Immunohistochemical and histopathological tests exhibited that galangin-loaded niosomes composed of non-ionic surfactant (Span 60, Span 40, and Span 20) and cholesterol demonstrated a significant decrease in neoplastic hepatic lesions and minichromosome maintenance 3 (MCM3) immunostaining hepatocytes related to hepatocellular carcinoma in rats. This nanoformulation-based galangin (10 mg), Span 60, cholesterol, and stearylamine had the diameter size range of 173.7-355.6 nm with drug entrapment efficiency and drug loading capacity of 77.69% and 16.72%, respectively [24]. Galangin and berberine inhibited oesophageal carcinoma cell growth and induced apoptosis. This combination treatment blocked the cell cycle at the G2/M phase and enhanced intracellular reactive oxygen species (ROS) levels. Combining these two treatments resulted in the downregulation of Bcl-2 and upregulation of Bax. In addition, *in vivo* study revealed that the combination treatment composed of galangin and berberine can significantly inhibit tumor growth without causing prominent toxicity [25]. Side effects of cyclophosphamide have led to clinical limitations of this chemotherapy agent. Galangin as a potential adjuvant agent can reduce hepatotoxicity resulting

from cyclophosphamide via up-regulation of Nrf2/HO-1 signaling (Figure 2).

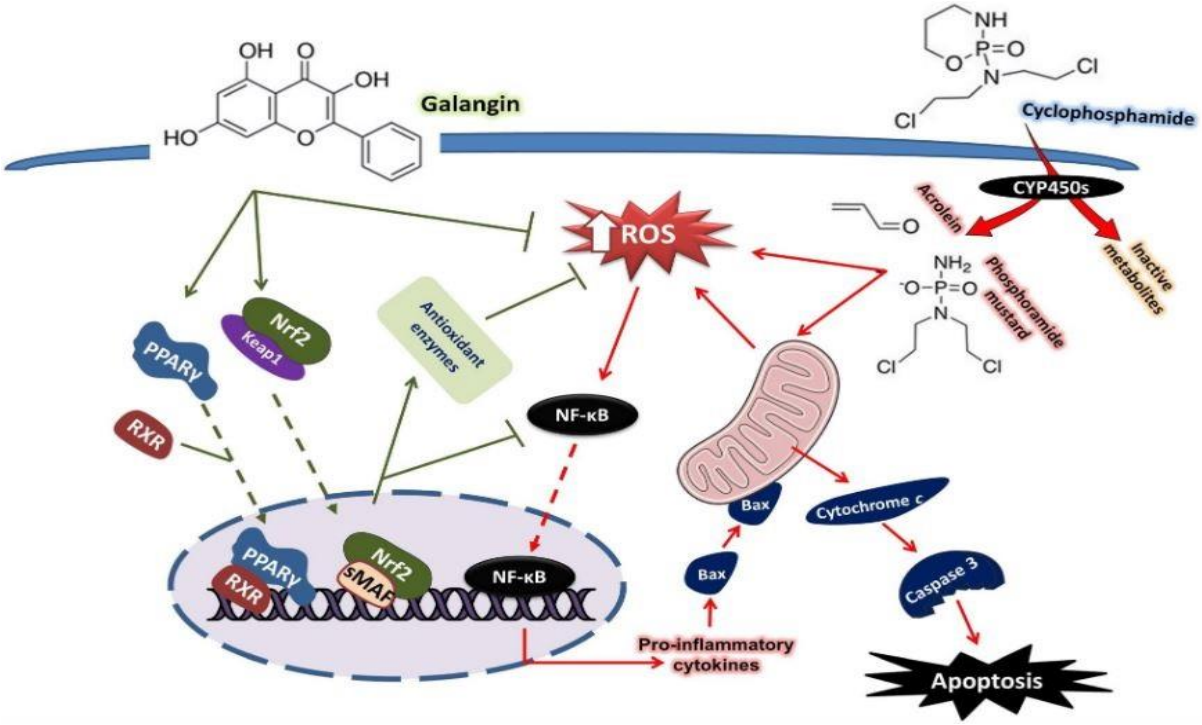

Fig. 2. Galangin prevents hepatotoxicity caused by cyclophosphamide via activation of Nrf2/HO-1 signaling (adopted from [26]).

Antimicrobial effect

The topoisomerase IV enzyme has a critical role in the severity of the antibacterial activity of galangin [27]. Galangin 3-methyl ether extracted from ethanolic and dichloromethane extracts of *Lychnophora markgravii* aerial parts exhibited antibacterial and antifungal activity against *Staphylococcus aureus*, *Escherichia coli*, *Pseudomonas aeruginosa*, and *Candida albicans* [6]. In a combination way, combinations of galangin, phenethyl caffeate, and chrysin showed the antibacterial effect on multidrug-resistant (MDR) microorganisms [28]. Molecular docking study revealed that galangin had higher binding energy of −51.36 kcal/mol compared to myricetin, quercetin, fisetin, hesperetin, naringenin, and hesperidin with binding energies of −57.19, −57.83, −60.65,−60.93, −64.90, and −69.31 kcal/mol, respectively [29]. Galangin, extracted from the shoots of *Helichrysum aureonitens* plant species, had *in vitro* antiviral effects on herpes simplex virus type 1 (HSV-1), coxsackievirus B type 1 (Cox B1), adenovirus type 31 (Ad-31), and reovirus. Galangin at concentrations ranging from 12–47 µg/mL had prominent antiviral impact on HSV-1 and Cox B1, but, limited antiviral activity toward reovirus, and no activity against Ad-31 [30]. Chrysin, pinocembrin, caffeic acid benzoic acid, p-coumaric acid, and galangin have been identified as the major component of ethanolic and aqueous propolis. Ethanolic and aqueous and extracts of propolis with different secondary metabolites such as galangin showed the 50% inhibitory concentration (IC_{50}) against HSV-1 plaque formation at 0.000035% and 0.0004%, respectively [31]. In another study, the antimicrobial activity of galangin was tested against the vancomycin-intermediate *Staphylococcus aureus* (VISA) strain Mu50. This study showed that the expression of the regulatory genes of murein hydrolase related to VISA strain Mu50 was hindered upon galangin treatment. Based on these findings, galangin may be useful as a promising antibacterial agent against infections resulting from VISA [32].

Anti-osteoporosis effect

In addition to suppressing the phosphorylation of p38 and extracellular signal-regulated kinase (ERK) of MAPK signaling pathway, nuclear factor of activated T cells 1 (NFATc1), C-Jun, and C-Fos protein by galangin, this metabolite reduces osteoclastogenesis in bone marrow–derived macrophages in a dose-

dependently way. In this regard, galangin can downregulate osteoclast-specific genes including lysosomal V0 subunit D2 (V-ATPase d2: vacuolar ATPase d2 related to the osteoclast plasma membrane pump), ATPase, tartrate-resistant acid phosphatase (TRAP), cathepsin K (CtsK), dendritic cell–specific transmembrane protein (DC-STAMP), and H^+ transporting [9].

Conclusions

Clinical limitations including sensitivity to light, temperature, pH, low insolubility in an aqueous medium, and semi-permeability to gastrointestinal barriers have been found for galangin. In the case of the anti-inflammatory effect, c-JNK, NF-κB, LPS-induced phosphorylation of p38 MAPK, and PI3K/Akt activity may be reduced by galangin. For anticancer activity, galangin can hinder metastasis by reducing the upregulation of MMP-2/-9. Moreover, the cell proliferation rate in human colorectal carcinoma cells has been reduced upon treatment by galangin in time-dependent way via blocking effect on HCT-116 clonogenicity and inducing cell cycle arrest at the G2/M or G1 phase. Galangin and berberine in a combination therapy inhibited oesophageal carcinoma cell growth by inducing apoptosis, blocking the cell cycle at the G2/M phase, and enhancing the ROS levels. In addition, galangin as a potential adjuvant agent attenuates hepatotoxicity caused by cyclophosphamide via up-regulation of Nrf2/HO-1 signaling. In the case of the antibacterial effect, the topoisomerase IV enzyme has a critical role in the severity of the antibacterial capacity of galangin. For anti-osteoporosis function, galangin down-regulates osteoclast-specific genes including V-ATPase d2 pump, ATPase, TRAP, CtsK, DC-STAMP, and H^+ transporting. Future investigations should be focused on novel micro and nano formulations to augment the bioavailability of this natural compound.

Study Highlights

- c-JNK, NF-κB, LPS-induced phosphorylation of p38 MAPK, and PI3K/Akt activity may be reduced by galangin.
- Galangin can hinder metastasis by reducing the upregulation of MMP-2/-9.
- The topoisomerase IV enzyme has a critical role in the severity of the antibacterial capacity of galangin.
- Galangin showed *in vitro* antiviral effects on HSV-1, Cox B1, Ad-31, and reovirus.
- For anti-osteoporosis function, galangin down-regulates osteoclast-specific genes such as V-ATPase d2.

Abbreviations

Ad-31: Adenovirus type 31
Akt: Protein kinase B
AP-1: Activator protein 1
c-JNK: c-Jun N-terminal kinase
Cox B1: Coxsackievirus B type 1
CREB: cAMP response element-binding protein
CtsK: Cathepsin K
DC-STAMP: Dendritic cell–specific transmembrane protein
ERK: Extracellular signal-regulated kinase
FoxO1: Forkhead box protein O1
HSV-1: Herpes simplex virus 1
HUVECs: Human umbilical vein endothelial cells
IC_{50}: Inhibitory concentration
IL: Interleukin
iNOS: Inducible nitric oxide synthase
LPS: Lipopolysaccharide
MAPK: Mitogen activated protein kinase
MCM3: Minichromosome maintenance 3
MDR: Multidrug-resistant
MMP-2/-9: Matrix metalloproteinase-2/-9
MMPs: Matrix metalloproteinases
mTOR: Mammalian target of rapamycin
NFATc1: Nuclear factor of activated T cells 1
NF-κB: Nuclear factor-kappa B
Nrf2: Nuclear factor-E2-related factor 2
PI3K: Phosphatidylinositol 3-kinase
PPAR-γ: Peroxisome proliferator-activated receptor-γ
ROS: Reactive oxygen species
TRAP: Tartrate-resistant acid phosphatase
V-ATPase d2: Vacuolar ATPase d2
VEGF: Vascular endothelial growth factor

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Conflict of interest

The authors declare that they have no conflict of interest.

Ethical approval

This article does not contain any studies with animals or human participants performed by any of the authors.

Author Contributions

All authors: conceptualization, preparing the first draft, and editing.

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