

# 硫酸多糖生物活性研究进展

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**摘要:** 硫酸多糖也被称为多糖的硫酸化衍生物,是硫酸根取代了多糖结构中单糖分子的部分羟基而形成的一类具有多种功能的物质,可以通过天然提取或硫酸化的结构修饰而得到。近年的研究证明硫酸多糖具有显著的抗肿瘤、抗病毒、抗氧化、抗凝血等活性,对机体免疫系统有一定调控作用。作者对硫酸多糖生物学功能的近期研究进行了总结,并对后续研究进行了展望。

**关键词:** 硫酸多糖; 抗病毒; 抗氧化; 抗肿瘤; 抗凝血; 免疫调节

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## Research Progress on Bioactivity of Sulfate Polysaccharide

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**Abstract:** Sulfated polysaccharides can be extracted from natural resources or modified from polysaccharides through substitution of the hydroxyl groups of monosaccharide molecules on polysaccharide chains by sulfate. The sulfated polysaccharides have multiple functions, such as anti-tumor, anti-virus, anti-oxidation, anti-coagulation and immunoregulation. In this paper, the recent studies on the biological functions of sulfated polysaccharides were summarized, and the future research was proposed.

**Keywords:** Sulfate polysaccharide, anti-virus, anti-oxidation, anti-tumor, anti-coagulation, immunoregulation

糖的生物学活性已为人们所共识。经过对糖生物学的多年不懈研究,人们已总结出一套应用于多糖的分离提取和结构解析高效的分析技术;同时对其抗氧化、抗感染、促进免疫等多方面功能和生物活性的研究也取得了较大进展<sup>[1-4]</sup>。多糖的生物活性

与其物理化学性质密切相关,如分子大小、单糖类型和比率以及糖苷结构特性等<sup>[5]</sup>。近年来多糖的化学分子修饰尤其是硫酸化修饰极大引起了研究者的兴趣。硫酸多糖是一类富含带有带负电荷硫酸根的化合物<sup>[6]</sup>,包括从动、植物中提取的各种天然硫酸多

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糖(如肝素),天然中性多糖的硫酸硫酸化修饰物及人工合成,半合成的各种硫酸多糖。硫酸多糖因具有广泛的生物学性质而受到人们的关注<sup>[7-8]</sup>。

## 1 硫酸多糖来源

### 1.1 天然硫酸多糖

硫酸多糖也称多糖硫酸酯或硫酸酯多糖,最先发现于软骨素以及从猪肠粘膜中提取的肝素,后来发现它还广泛存在于海藻类植物。肝素是动物体内一种天然抗凝血物质,临床上有广泛应用<sup>[9-10]</sup>。

海藻中红藻、褐藻、绿藻等中含有丰富的硫酸多糖。红藻细胞壁上提取的卡拉胶<sup>[11]</sup>、海洋褐藻和棘皮动物中提取的糖硫酸酯、绿藻提取物硫酸化半乳糖<sup>[12]</sup>具有多种生物活性如抗病毒、抗肿瘤、抗凝血等而无毒副作用,可用于食品、药物及保健品中<sup>[13]</sup>。

### 1.2 硫酸化修饰多糖

目前制备硫酸化多糖常用方法包括浓硫酸法、氯磺酸-吡啶法、三氧化硫-吡啶法、氯磺酸-甲酰胺法等<sup>[7,14]</sup>。硫酸基团的引入对多糖多种生物活性有影响。糖分子自由-OH被-SO<sub>3</sub>H取代后,使多糖的电荷和静电作用会增加,从而增强水溶性和构象的转变。Zhang等<sup>[15]</sup>通过氯磺酸-吡啶法修饰灵芝多糖,检测其物理化学性质、生物学功能的变化,发现其水溶性随取代度的增加而增加,并且提高了其抗氧化和抗肿瘤活性。多糖经硫酸化后具有多方面的生

物活性或者是增强了原来多糖的生物活性,这与它的硫酸根取代度、相对分子质量、结构转变有关<sup>[16]</sup>。

## 2 硫酸多糖生物活性

### 2.1 抗病毒

1958年, Gerber<sup>[17]</sup>首次报道硫酸多糖的抗病毒活性,他们研究发现从石花菜中提取的硫酸多糖 cartilagenium 能预防鸡胚被流感病毒 B 和腮腺炎病毒感染。许多种类的海藻含有大量的结构复杂的硫酸多糖已被证明抑制包膜病毒的复制,包括 DNA、RNA 病毒等(见表 1)。川明参多糖是一种天然的硫酸多糖,在 77.12~104.81 μg/mL 质量浓度范围内显著抑制鸭肠炎病毒(Duck enteritis virus, DEV)的活性<sup>[18]</sup>。

木耳(*Auricularia auricula*)在亚洲具有悠久的历史,被广泛用于医药和保健品中。木耳重要活性成分多糖(AAP)具有生物学功能。Nguyen 等人<sup>[28]</sup>研究发现木耳多糖硫酸化修饰后比修饰前相比抗病毒活性增强。硫酸多糖主要是作用于病毒囊膜蛋白,从而阻止病毒感染早期阶段,包括初始吸附到靶细胞以及病毒的侵入<sup>[29]</sup>。

### 2.2 抗氧化

活性氧(ROS)攻击细胞膜大分子蛋白质和 DNA,导致严重的组织损伤性疾病,如癌症、糖尿病、神经退行性疾病和炎症。抗氧化剂可以减缓活

表 1 从不同的海藻中提取的硫酸多糖的抗病毒活性

Table 1 Antiviral activity of sulfated polysaccharides from different seaweeds

来源	主要成分	病毒类型	IC <sub>50</sub> (μg/mL)	参考文献
红藻类				
<i>Cryptonemia seminervis</i>	DL-galactan	HMPV	12.5~100	[19]
<i>Scinaia hatei</i>	xylomannans	DENV	1.1	[20]
<i>Gracilaria corticata</i>	galactan	DENV	10.0	[20]
<i>Lithothamnion muelleri</i>	heterosaccharide	HSV-1, HSV-2		[21]
<i>Nemalion helminthoides</i>	Mannose	HSV-1	5.4	[22]
褐藻类				
<i>Sargassum patens</i>	D-galactose	HSV-1	1.5	[23]
<i>Sphacelaria indica</i>	Xylogalactofucan, Sea oxalic acid	HSV-1		[24]
<i>Adenocystis utricularis</i>	Fucoidan	HIV-1	0.6~70	[25]
<i>Laminaria angustata</i>	xylogalactofucan	HSV-1	0.2~25	[26]
绿藻类				
<i>Caulerpa racemosa</i>	heterosaccharide	DENV	0.6	[20]
<i>Codium fragile</i>	Galactose	HSV-2	4.7	[27]

性氧,如OH、H<sub>2</sub>O<sub>2</sub>和O<sup>2</sup>引起的氧化应激过程,有益于人体健康<sup>[30]</sup>。许多合成的抗氧化剂在食品和医药行业中被广泛应用,如二叔丁基对甲酚、叔丁基羟基茴香醚(BHA)、特丁基对苯二酚(TBHQ)和没食子酸丙酯(PG)等。然而,这些合成的抗氧化剂具有一定的毒副作用<sup>[31-32]</sup>。天然抗氧化剂具有不可替代的优势,如硫酸多糖等。已有文献报道,江蓠(*Gracilaria debilis*)<sup>[33]</sup>、毛茛(*Pterocladia capillacea*)<sup>[34]</sup>、裂片石蓴(*Ulva fasciata*)<sup>[35]</sup>以及肠浒苔(*Ulva intestinalis*)热水提取物中的褐藻素、海带多糖、海藻酸等成分具有很强的清除DPPH、O<sup>2</sup>·、OH、H<sub>2</sub>O<sub>2</sub>的活性,是良好的抗氧化剂。Yuan等<sup>[36]</sup>采用微波提取技术从球型褐藻中提取其硫酸多糖,发现在不同温度水浴条件下提取的岩藻糖(fucose)和葡萄糖醛酸(glucuronic acid)都具有清除DPPH和还原能力。

党参多糖(CP)是党参(*Codonopsis pilosula*)重要的活性成分,具有药理活性。Liu等人<sup>[37]</sup>采用氯磺酸一吡啶法对其进行酯化改性,提高了其清除自由基活性。体内实验结果显示,硫酸化党参多糖(sCP)能显著降低肝损伤模型小鼠血液中谷丙转氨酶(ALT)、谷草转氨酶(AST)、肿瘤坏死因子- $\alpha$ (TNF- $\alpha$ )和肝组织丙二醛(MDA)的含量,并且肝组织中SOD、GSH-Px显著高于模型组和党参多糖组。结果表明,天然多糖经过硫酸化修饰后其抗氧化活性会发生不同程度的增强。

### 2.3 抗肿瘤

目前恶性肿瘤已被认为是危害人类健康的疾病之一。天然硫酸多糖已被证明具有较强的抗肿瘤活性,其抗肿瘤活性主要表现在抑制肿瘤细胞的增殖,促进肿瘤细胞凋亡以及通过提高免疫系统的功能来协同杀伤肿瘤细胞。马尾藻(*Sargassum horneri*)水溶性硫酸多糖SHP30能将人结肠癌上皮细胞(DLD)阻止在G<sub>0</sub>/G<sub>1</sub>期,从而抑制其生长;实时荧光PCR结果显示,它降低了Bcl-2的表达,增加了Bax的表达<sup>[38]</sup>。已有的研究发现硫酸化修饰可在一定程度上提高多糖抗肿瘤活性。O-硫酸化大肠杆菌K5荚膜多糖(OS-K5PS)能抑制纤维母细胞生长因子(fibroblast growth factor, FGF)的表达而抑制乳腺癌细胞S115的增殖,而NS、OS-K5PS没有这种活性<sup>[39]</sup>。然而,Teng等报道了NS、OS-K5PS显著抑制SDF-1/CXCL12趋化因子诱导的B16黑色素瘤细胞的增殖,使细胞周期停滞在G<sub>0</sub>/G<sub>1</sub>期,而且其抑制

效果高于OS-K5PS<sup>[40]</sup>。另一研究发现,NS、OS-K5PS能抑制转化生长因子- $\beta$ 1(transforming growth factor- $\beta$ , TGF- $\beta$ )诱导的骨转移性乳腺癌细胞MDA-MB-231中溶骨因子IL-11的表达,小鼠模型证明NS、OS-K5PS抑制了肿瘤的生长和癌细胞的转移<sup>[41]</sup>。硫酸多糖不同的生物活性可能与硫酸取代度、硫酸化作用的位点密切相关。

### 2.4 抗凝血

肝素是最早应用于临床的天然抗凝剂,它具有相对分子质量小、抗血栓作用强、口服易吸收、体内半衰期长、生物利用度高等优点,有望成为安全、高效的抗血栓类药物<sup>[42-43]</sup>。海藻类富含硫酸多糖,也是天然抗凝剂的丰富来源<sup>[44]</sup>。松藻(*Codium divaricatum*)<sup>[45]</sup>及礁膜(*Monostroma oxyspermum*)<sup>[46]</sup>硫酸多糖能剂量依赖性地延长凝血酶活化时间。因此,天然硫酸多糖在预防及治疗由凝血造成的心血管疾病(包括心肌梗塞、中风、深静脉血栓形成等)方面具有巨大的应用潜力。

### 2.5 免疫调节

研究表明,硫酸多糖具有免疫调节作用,包括免疫促进作用和免疫抑制作用。Cao等人<sup>[47]</sup>从昆布(*Ecklonia cava*)水提取物中通过阴离子交换色谱分离纯化得到其硫酸多糖,并检测其免疫增强作用。Western blot结果显示,昆布硫酸多糖通过MAPK和NF- $\kappa$ B信号通路促进RAW264.7细胞产生一氧化氮(nitric oxide, NO)和促炎因子。从酿酒酵母(*saccharomyces cerevisiae*)得到的硫酸葡聚糖单独免疫小鸡后增强了小鸡脾脏中免疫细胞的分化,与新城疫疫苗同时免疫小鸡后42d,能显著提高血液中抗体滴度和细胞因子白介素-2(interleukin-2, IL-2)和 $\gamma$ -干扰素(interferon- $\gamma$ , IFN- $\gamma$ )的表达水平<sup>[48]</sup>。然而,紫菜(*Porphyra haitanensis*)多糖能抑制过敏性小鼠血清中IgG、IgG1以及IgE水平,降低脾脏细胞中Th2型细胞因子(IL-4、IL-5和IL-13)的表达,增强了Th1型细胞因子IFN- $\gamma$ 的表达<sup>[49]</sup>。

研究发现,适当的分子修饰或结构改变使多糖产生新的活性或进一步增强原有的功能,尤其是硫酸化修饰。硫酸修饰会显著提高多糖的免疫调节作用,如增强免疫细胞的增殖,促进细胞因子的产生,作为佐剂增强疫苗免疫反应或抑制促炎因子的产生而抑制炎症的发生等(见表2)。

表 2 硫酸化修饰多糖的免疫调节活性

Table 2 Immune regulate activity of sulfated polysaccharides

生物学活性	来源	硫酸化修饰方法	参考文献
抗炎作用	<i>Antrodia cinnamomea</i>	维素硫酸酯化	[50]
	<i>Armillariella mellea</i>	维维素硫酸酯化	[51]
	<i>Astragalus polysaccharide</i>	氯磺酸-吡啶法	[52-53]
	<i>Radix Astragali</i>	氯磺酸-吡啶法	[54]
免疫增强作用	<i>Ganoderma atrum</i>	氯磺酸-吡啶法	[55]
	<i>Ganoderma lucidum</i>	硫磺三氧三甲胺	[56]
	<i>Bush Sophora</i>	氯磺酸-吡啶法	[57]
	<i>Poria cocos</i>	氯磺酸-吡啶法	[58]
疫苗佐剂作用	<i>Astragalus polysaccharide</i>	氯磺酸-吡啶法	[59]
	<i>Auricularia auricula</i>	氯磺酸-吡啶法	[60]
	<i>Tremella, Codonopsis pilosula</i>	氯磺酸-吡啶法	[61]
	<i>Ophiopogon japonicus, Fructus jujubae</i>	氯磺酸-吡啶法	[62]

### 3 展望

天然来源的产物在医药、营养食品、药用化妆品和功能性食物等领域研究取得了多方面的进展,如动、植物、微生物来源的多糖,尤其是硫酸多糖,具有多种生物活性<sup>[63]</sup>。最近的研究表明,天然多糖经人工硫酸化修饰后不仅可以增强其原有的生物活性,还可产生新的生物活性。硫酸多糖在医药、营养食品、药用化妆品和功能性食物等领域具有广阔的应用前景(图 1)。目前使用的多糖硫酸化修饰方法包括氯磺酸-吡啶法、三氧化硫-吡啶法、浓硫酸法

等,使用的都是强酸、强氧化、强腐蚀性溶剂,如果反应时间过长,易引起多糖降解,导致原有活性丢失。需要对反应体系进一步优化(如加入催化剂),提高反应速率,降低多糖降解率。多糖硫酸化修饰后的活性与其水溶性、相对分子质量、硫酸基团取代度、硫酸基团的位置及多糖结构变化等具有相关性。因此,需要对多糖硫酸化的工艺进行优化,包括反应时间、硫酸化试剂、溶剂与多糖溶解度、调节剂等,明确不同工艺修饰后硫酸多糖的结构变化,检测硫酸多糖结构与功效之间的关系,建立质量控制标准;同时需要加强硫酸多糖的毒副作用相关研究,为其临床应用奠定基础。

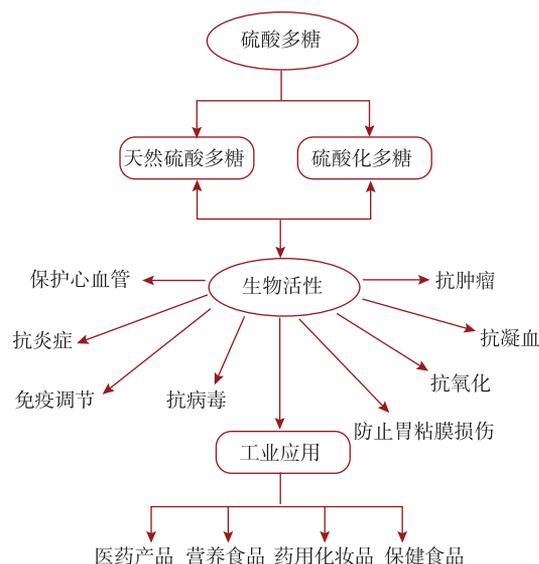


图 1 硫酸多糖的生物学特性和潜在的工业用途

Fig. 1 Biological properties and potential industrial uses of Sulfate polysaccharide

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## 会 议 消 息

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主办方: 中国化学会

承办方: 1、中国化学会有机化学学科委员会; 2、南方科技大学; 3、中科院上海有机化学研究所;

会议主题: 自由基与催化

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会议内容: 展示近年来国内在有机自由基化学领域取得的研究成果, 促进有机自由基化学及相关学科的发展壮大。主要交流内容: 1、有机自由基反应; 2、有机光催化; 3、有机电化学; 4、不对称自由基反应; 5、自由基在合成中的应用; 6、自由基机理研究。