



## **Conifer Cones: An Alternative Raw Material for Industry**

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### **Authors' contributions**

*This work was carried out in collaboration between both authors. Author HTS designed the study, managed the literature search and wrote the first draft of the manuscript. Author OUY performed the analyses of the study and the literature searches. Both authors read and approved the final manuscript.*

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### **ABSTRACT**

The cone is an organ of conifers that contains the reproductive structures. There is growing interest products from natural materials in worldwide. However, the chemical compounds from cones are great interest because of its residue products. A number of studies on conifer cones have available in literature. Majority of these studies are on morphological, chemical and material properties of cones.

All these findings have contribute to better utilization of materials from conifer cones as new medicinal plant products. However, it was consistently reported that the chemical constituents of cones changed depend on geographical, seasonal, genotypic and environmental situations. It was reported that conifer cones have similar chemical constituents like wood but in various proportion. They markedly contain rich of phenolic compounds and some extractives. They primarily consists cellulose, lignin, and hemicelluloses as major constituents. They have also some condense tannins, resin acids, stilbenes, flavonoids, etc. Many of researchers have already reported that these extracts, which have various proportions in different species may offer some advantages in terms of utilization from waste materials and may show exceptional medicinal properties.

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A number of different approaches have applied to find alternative and economical utilization from coniferous cones. It has also used for manufacturing various kinds of composite and paper material as substituent of wood. It has already tried to use for particleboard, fiberboard and plastic composite process aiming to without lowering properties. The results reported in that area is promising. It has been utilized for purifying of waters as bioabsorbent or scavengers for heavy metals from waste streams. It has also found to be useful for producing cellulose with its intrinsic viscosity and molecular weight. However, it has already well explained that some extracts from cones show antioxidant, antifungal, antimicrobial properties and may cure some disorders of humans. The utilization of these constituents from as a forest residue material may offer many possible applications as raw material.

**Keywords:** *Conifer tree; cone; cellulose; extractives; antioxidant; polysaccharides; antifungal; stilbenes.*

## 1. INTRODUCTION

Trees and its components such as; barks, needles, leafs, branches have being used from the dawn of civilization. However, million tonnes of logging residues such as; barks, cones, needles, twigs, and etc. have generated during timber harvesting in forestlands. These residues differ considerably in volume per acre, size and location. Moreover, scientists and engineers have investigated thoroughly a wide variety of ways to manipulate lignocellulose based materials at the chemical and physical level to take advantage of their enhanced properties and greater useability, as compared with their counterparts.

The trees from *Pinaceae* family (i.e. pinus, abies, picea, cedrus, latrix, etc.) are one of the most widely distributed in worldwide, with approximately 250 species in 11 genera [1]. The majority of the species have been in temperate climates, but ranging from subarctic to tropical lands. However, due to their high tolerance to dryness, and chilly wheather, many pinaceae species have been planted for protection against tides, producing fuels and substitution of wood based materials [2].

The cone is an organ of conifers that contains the reproductive structures. Cone development, with viable seed, occurs normally in 15- to 20-year stands, and persists until well past maturity (up to 100 years). However, the cone yield decreases considerably as tree become older [3]. Its structure varies markedly between the different species (i.e. conic, cylindrical, ovoid, small to large, short to long, etc.) and is often crucial for the identification of species of conifers. But climatic regimes has also influenced seeding habits and dispersal patterns while close relationship between cone size, number of

scales, and the average number of seed per cone [3-5]. The cones are an important indication of fire risk for land managements both as fire hazard and refrostration difficulties. These residues could be creates conditions not suited to germination by blocking soil and minimizing moisture and light [2,5].

There is growing interest in medicinal products from natural materials worldwide. However, the extracts from cones are great interest because of its residue products and not utilized properly. Hence, a significant portion of recent literature on cones have focused on chemical analysis with the goal of determining and isolating valuable compounds. These extracts, which have various proportions in different species may offer some advantages in terms of utilization from waste materials and may show exceptional medicinal properties.

## 2. CONE MORPHOLOGY OF CONIFERS

There are two types of cone one called *female cone*, which produces seeds and other *male cones*, which produce pollen. The male cones are small, typically one to five centimeters long, and only present for a short period, falling as soon as they have shed their pollen. However, the female cone contains ovules that when fertilized by pollen, become seeds. The ovules develop into seeds after fertilization by pollen grains. The bract scales develop first, and the seed scales develop later to enclose and protect the seeds. Maturation takes 6–8 months from pollination in most *Pinaceae* genera, but typically 12 months in cedars and 18–24 months in most pine species [1-5].

Typically, a pine cone could go through many cycles of opening and closing during its life span, even after seed dispersal is

complete. Figs. 1 and 2 show a typical cycle and lifespan of pine cone, respectively [1-4]. However, the condition of fallen pine cones is a crude indication of the forest floor's moisture content. Closed cones indicate damp conditions while open cones indicate the forest floor is dry.

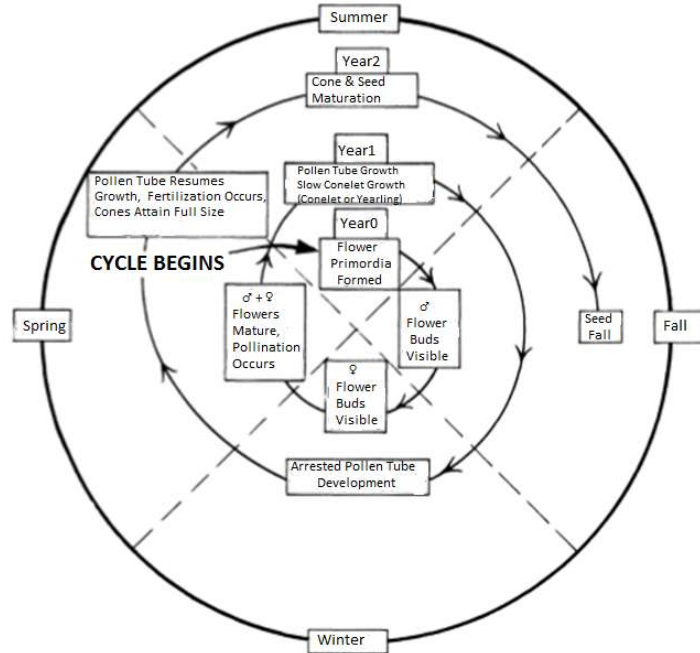


Fig. 1. Typical cycle of cones [4]

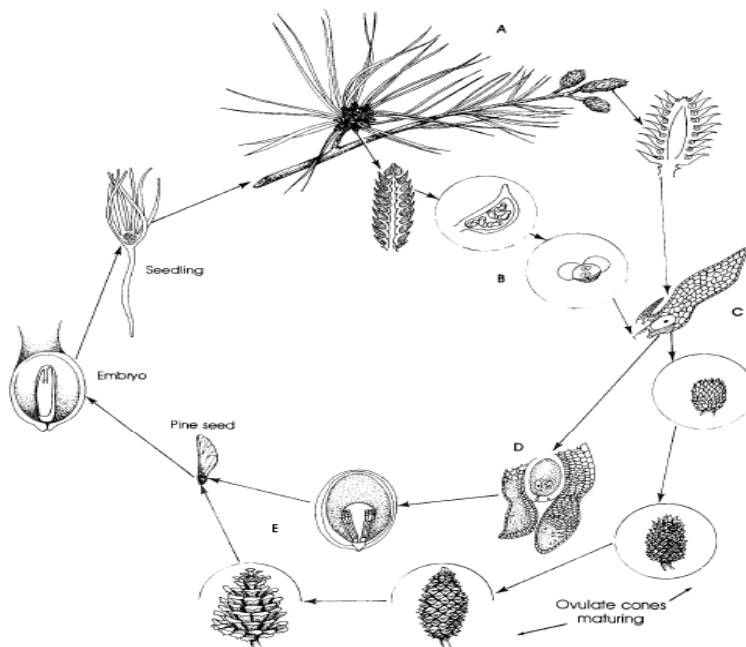


Fig. 2. Lifespan of a pine cone (A: At maturity, male and female cones produces by tree, B: Pollen produced by male cones, C: Blown inside the female cones, D: Pollen tube ghrowth and fertilization leads to produce zygote, E: Following mitosis an embryo, a new seed developed within female cone) [2,4,5]

Although time requirement varies between species, the cones have usually take two or three years to mature. However, juvenile cones may have a distinctive color, but normally identification requires mature cones. The portion of the cone scale that is exposed before the mature cone opens is thickened and is called the *umbro*. The cone scales may remain closed and sealed by resin long after cone maturity. Typically, cones open at maturity and release seed. The seed may be wind-carried, in which case it is normally small and light with a wing longer than the seed; or it may be dispersed by animals.

### 3. CHEMICAL ANALYSES OF CONES

Researchers have already reported some valuable informations for chemical composition of cones of coniferous tree, and isolated extracts [6-10]. They have usually compare their results with various type of cones and other studies in order to find differences in chemical contents. All these information have contribute to better utilization of materials from coniferous cones as novel pharmaceutical plant products. However, it was consistently reported that the chemical constituents changed depend on geographical, seasonal, genotypic and environmental situations [6-10].

However, a number of analytical methods used to determine the monosaccharide composition of polysaccharides have been reported, such as; gas chromatography (GC) and high-performance liquid chromatography (HPLC) [11-13]. The GC which is a well established technique, has been widely used for carbohydrate profiling of lignocellulosic materials, especially for detecting carbohydrate monomers in complex structures [12]. Although GC is a sensitive technique, a derivatization methods before GC analysis are needed to increase volatility of monosaccharides and decrease interaction with the analytical system. These are mainly including trimethylsilyl (TMS), aldonitrile acetate and alditol acetate procedures [11-14]. But the TMS is usually preferred due to determine complex chromatographic patterns owing to anomerization, whereby the  $\alpha$ - and  $\beta$ - anomers of the pyranoside and furanoside forms of the monosaccharide can produce multiple peaks [11,12].

However, aldonitrile acetate and alditol acetate methods have been usually preferred for monosaccharide component analysis in both

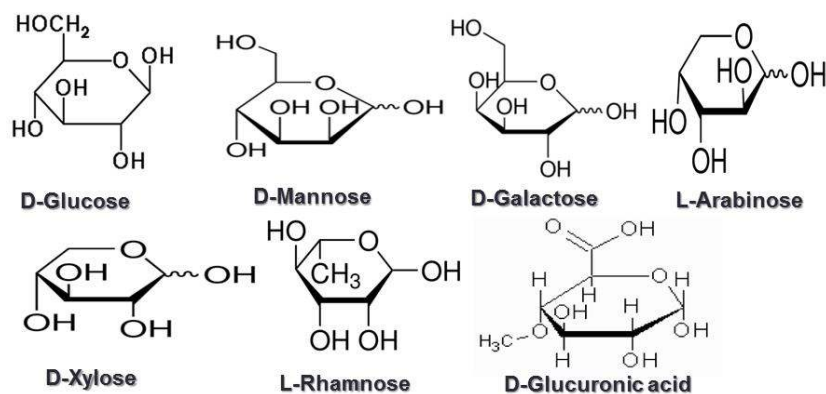
neutral and amino sugars. Alditol acetates can be formed in a one-step reaction and, once formed, are stable allowing post-derivatization cleanup and storage of treated samples for extended periods than aldonitrile acetate which is usually formed by two-step derivatization procedure and lack of reproducibility for some amino sugars. Moreover, acetylation of alditols eliminates the anomeric center and therefore simplifies the chromatograms, as most sugars produce one chromatographic peak. Due to complex chemical structure and great variations in constituents, some modified approaches derived from traditional wood chemistry techniques, applied to determine chemical constituents of cones [8-16].

#### 3.1 Chemical Constituents of Cones

It was reported by a number of researchers that coniferous cones have similar chemical constituents like wood but in various proportion. But they markedly contain rich of phenoloic compounds. They typically consists cellulose, lignin, and hemicelluloses as major constituents. It has already found that various compounds can be extracted from pine cones, such as polysaccharides, lignin-related compounds, water and solvent soluble substances and essential oils, among which the content of polysaccharide accounts for about 50% (w/w) [7]. It was proposed that the *pinus* and *picea* cone polysaccharides typically consisted of ribose, rhamnose, arabinose, xylose, mannose, glucose and galactose in different molar ratios [8-10,14-17]. The typical polysaccharides found in pine cones shown in Fig. 3.

However, cones from some *pinus* and *picea* species have considerably higher condense tannins [8]. Eberhardt et al. [9] have also proposed that extracts from *Picea glauca*, *pinus ponderosa* ve *pinus banksiana* cones had rich resin acids and shows hydrophobic properties. It was found that *pinus nigra* cones constituents approx. 65.7% polysaccharide, 26.7% klason lignin, and around 10% extractive compounds. It was reported by Font and his group [18] for mediterranean pine cone that it contains 7.3% ethanol extracts, 21.7% hemicellulose, 39.3 cellulose and 30.9% lignin.

Moreover, for lignin, the chemical groups found in cone similar to coniferous woods but rich in phenylpropane groups [19]. Table 1 shows polysaccharide composition of some conifer seed cones [20].



**Fig. 3. Typical polysaccharides in pine cones**

In a study, a total of 57 compounds have been identified in the essential oil from *Chinese White Pine* (*Pinus armandii*) cones. They have mainly;  $\alpha$ -pinene, *D*-limonene,  $\beta$ -pinene and *trans*-pinocarveol (4.76%) and less amount other compounds [17]. In other study on same species, high amounts of *terpenoid* diversity and variations have been detected [16]. However, terpenoids represent a mixture of volatile *monoterpenes* (C<sub>10</sub>) and *sesquiterpenes* (C<sub>15</sub>), which solvate higher molecular weight *diterpenes* (C<sub>20</sub>), called as *oleoresins*.

However, *stilbenes* which is small family of plant secondary metabolites, phenolic extractives found in cones. The stilbenes have plant disease resistance while are considered as fungicide properties. They have been synthesized when biotic and abiotic stresses occur. They protect the plant from infection and subsequent decay. They are most often two aromatic rings, contains double bond linkage and naturally present most often in *trans* form. Moreover, stilbenes differ on ring substitution. Most stilbenes possess resorcinol group but some have ethers and a few form glycosides [12,13]. Fig. 4 shows some of the chemical structures of stilbenes.

#### 4. UTILIZATION OF PINE CONES

Although pine cones have received considerable attention for value added products, but the residual cone data is non-existent. However, the annual residue of pine cones was probably millions metric tonnes per year worldwide. Because of their widespread occurrence, cones have been a traditional part of the arts and crafts of cultures where conifers are common. Examples of their use includes seasonal wreaths

and decorations, fire starters, bird feeders, toys [5]. Moreover, particularly fundamental work carried on the understanding of basic chemistry of extractives and their possible isolation and derivatization to pharmaceutical products [6,7,15-17,21,22].

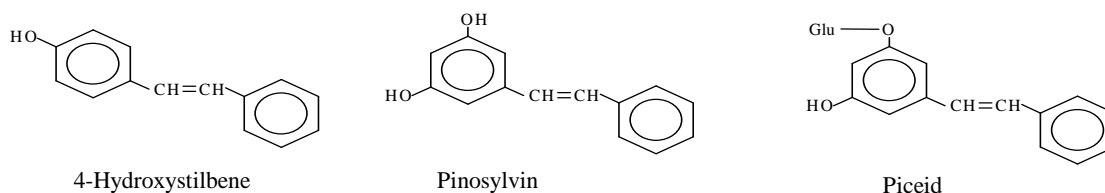
Biomass energy is one of humanity's earliest sources of energy particularly in rural areas where it is often the only accessible and affordable source of energy. Worldwide biomass providing very high proportions of the world's energy needs of all human and industrial processes. The cones from coniferous trees could be potential utilized as energy resource. The energy value of red pine (*pinus buritita*) cones indicates approximately 10% lower than red pine wood energy level (4350 vs 4800 kCal/kg) [18]. But this level of utilization have considered to be low value from lignocellulosic materials. This burning has also produce wastes, that is, normally unused and undesirable products of a specific process. It was found that typically, a pine contains, 54.2% carbon, 6.8% hydrogen, 0.3% nitrogen and 0.8% ash content. However, the final residue of burning is different in various conditions depending on the temperature, indicating that there are some fractions that decompose at high temperatures.

The chemical constituents of plants usually effects their potential medicinal properties. Especially quantity and quality of essential oils, phenolic substances and other extractives are important parameters for utilization of that as antioxidant, antimicrobial, or medicinal substances.

Because plants grows (conifers) varies in climate and elevation, chemical distribution is also

**Table 1. Polysaccharide composition of some conifer seed cones [20]**

Sample	Arabinose	Galactose	Glucose	Xylose	Mannose
<i>Picea glauca</i>	4.5	6.6	52.5	10.2	26.2
<i>Pinus banksiana</i>	2.7	4.8	58.8	5.5	28.2
<i>Pinus nigra</i>	4.0	4.6	58.7	10.1	22.6
<i>Pinus ponderosa</i>	4.5	4.8	56.6	10.0	24.1
<i>Pinus resinosa</i>	4.0	4.8	61.1	9.7	20.4
<i>Pinus taeda</i>	4.2	5.8	60.2	11.1	18.7

**Fig. 4. Some stilbenes naturally occur in pine cones**

varied. The literature has clearly explained that chemicals from cones, in general, acts as both a chemical defense, because of the activity of single constituents and a physical defense, because of its ability to expel some herbivores with resin pressure and to crystallize and protect wound sites. To date, various pharmaceutical compounds have been found and isolated from coniferous cones.

It has already reported by a number of scientists that chemicals from cones could be an alternative medicine producing method compared to conventional processes. On the other hand, one approach for possibly *antioxidant* and *antifungal* effects that has received increased attention in recent years has the idea of using cone extracts as medicine, fungicide or insecticide [15-17,20-22].

However, pine cones are rich in terpenoids that represent the primary chemical defense in conifers, occurring as oleoresin. These compounds possibly useful as fragrances in cosmetics, flavoring additives for food and beverages, scenting agents in a variety of household products, and intermediates in the synthesis of perfume chemicals. They are also used for medicinal purposes in aroma therapy as carminative, rubefacient, and abortifacient agents [15,17]. Terpenoids have been also utilized in ethnomedical practice throughout the World such as; cure diarrhea, in cough remedies, help break fever [17,20]. However, the ratio of lower molecular weight mono terpenes (and some sesquiterpenes) to diterpenes determines the viscosity of oleoresin. This physical property

dictates how resin flows within a plant and, as a result, the ability of an internal parasite to move through it or the capacity of an external herbivore to eat tissue that contains it [18].

Proanthocyanidins (flavonoids) are also phenolic compounds and could be found in many tree's barks, fruits and leaves. This phytochemical typically inhibits microorganisms (fungi, bacteria and viruses). They produce some complexes with cell enzymes to resist against fungal attacks or encapsulated cell wall polymers hence protect against disruptive enzymes from out of cell wall [21,12]. These show some evidence for antioxidant activity of extracts from pine cones [8].

Researchers have already reported that extracts from pine cones have antioxidant activity and proposing for curing some human disorders [7-11]. It was found that polysaccharides extracted from pine cones, which have the potential to be used as novel antioxidants. They showed various pharmacological activities, such as anti-tumor, anti-microbial, anti-HIV, properties [6-10,16,21].

Celimene et al. [21] reported that pine cones contains stilbenes that particularly pinosylvin which has demonstrated anti-fungal activity. However, the pinosylvin has well known active agents in many herbal medicines and already patented as *anti-microbial agent*. It is a crystalline product and insoluble in water but soluble in ether and alcohol. Moreover, the monomethyl and dimethyl ethers also serve as *anti-fungal agents* and typically found in cones. It was reported that the toxic properties of stilbenes

are based on the interactivation of fungal enzymes which contain *-SH groups* in their active sites [21-23].

It was established that phenolic compounds are rich in pine cones tend to have decay resistance properties. However, resin acids and tannins obtained from pine cones have potential applications as preservative and antifungal agents [7,9,24]. They proposed that due to variations in lignin structure and rich phenyl propane units, it may useful for cure HIV viruses [19]. Extracts from cone of *Pinus glauca*, *Pinus ponderosa* ve *Pinus banksiana* have found to be effective particularly on *Brown-rot fungi* [21].

It was reported by a number of researchers that cones have considerably higher phenolic compounds. However, it is important to mention that depend on age, cone can contain different groups in various properties. It was found that the extracts from same species but different parts of tree have shown different *anti-oxidant* activity. It was ordered as effective levels from less to highest as; pine wood < mature pine cone < pine bark < fresh pine cone < pine needle < juvenile pine cone [24].

Sabharwal [25]. found that pine cones could be useful for producing cellulose with its intrinsic viscosity and molecular weight. He also proposed that carboxyl methyl cellulose (CMC) could be prepared from ground pine cones and would meet the commercial specifications of some of the CMC preparations. Cellulose is cheap, hydrophilic, chemically modifiable and non-toxic substance that some cellulose derivatives can be utilized in various applications such as; bio-medical science.

The utilization of pine cone as purifying waste streams or as biosorbent have also been investigated. Demirak et al. [26] reported that the surface of pine cone powder could be modified by sodium hydroxide solution and the chemical composition of pine cone powder samples is changed after biosorption of ammonium. This forms an interesting option for wastewater treatment, as a possible non-conventional biosorbent for the removal of ammonium. However, the pine cones could be utilized as scavengers for heavy metals from waste streams [25].

However, the use of cones in composite and pulp industry as raw materials has been considered but the logistic of tonnage, collection and delivery problems may be the major obstacle for its

potential use in such industries. Due to low density and very short fiber length may render his material. In a estimation, the packing density of the cones was calculated approx  $100 \text{ kg/m}^3$  [25].

Pine cones are biomass that could be of use as composites manufacturing (i.e. particleboard, fiber board and plastic composites) itself or wood substitute. The physical and chemical properties of pine cone was the subject of interest of research, focusing on some aspects considered as follows; there are some studies concerning the influence of additives on the particleboards. Sahin and Arslan [27] tested the influence of pine cone particles in red pine (*Pinus brutia*) wood particles for particleboards at outdoor conditions. They found that pine cone particles effects improving weathering performance of panels at various level. It was also determined that the *Pinus pinea* cone flours could be used as wood substitute during medium density fiberboard manufacturing at acceptable level of strength properties [28]. Ayilmis et al. [29] have used the pine cone flour as reinforced material in polypropylene based plastic composite panel manufacturing (WPC) and determined the optimum conditions for the mixture. The results shows that the flexural properties and water resistance of the WPC samples were not significantly affected by the addition of 10% (by weight) of the cone flour when compared to the WPC samples made from only wood flour. In another study, the particles from cone of the stone pine can be considered as an alternative to wood material in the manufacture of particleboard used in indoor environment due to lower thickness swelling, water absorption and lowering formaldehyde emission [30]. These are well examples of pine cones that could be useful in wood based composite industry as wood substitute material at some level.

The kraft pulping properties of *European black pine (Pinus nigra)* cones alone and with same wood chips were evaluated by Gulsoy and Ozturk [31]. It was found that when the ratio of cone chips was increased, resulting higher kappa numbers but lower total screened yield of pulps. However, the strength and brightness of pulps were decreased gradually when the cone fibers used in cooking proportions. But it was proposed that it is possible to utilize cone particles with wood chips in Kraft pulping at optimum level to find acceptable strength and physical properties of pulps.

## 5. CONCLUSIONS

Extracts from different parts of conifer species (i.e. bark, needle, cone and resin) have been utilized in many areas for their specific applications. However, these are typically lignocellulosic materials that are composed of cellulose, hemicellulose, lignin and extractives in varying proportions. Moreover, utilization of these constituents offer many possible applications as raw materials. It has well established that phenolic compounds, resin acids, tannins, stilbenes, which are rich in pine cones, possess antioxidant and antifungal properties. But it is important to mention that depend on age, geographic region, species of cone could be contain different groups in various properties.

However, the use of cones in composite and pulp industry as raw materials could be difficult due to collection and delivery problems. But it is potentially looks useful for utilization of chemical industry as raw material for manufacturing some pharmaceutical substances.

## CONSENT

It is not applicable.

## ETHICAL APPROVAL

It is not applicable.

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## COMPETING INTERESTS

Authors have declared that no competing interests exist.

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