Effects of oak wood on the maturation of alcoholic beverages with particular reference to whisky

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Summary

Oak casks are used for the maturation of a wide range of alcoholic beverages. Focusing on whisky production, this paper reviews the influence of oak wood properties on the flavour of alcoholic beverages. It examines whether the selection of wood or casks on the basis of their effects on flavour can be justified given our present understanding of the process. The current use of oak casks in whisky manufacture is briefly summarized and the wood properties, both chemical and anatomical, that might influence flavour are described. These characters vary in both the virgin wood and the used casks. The factors influencing this variation are identified. The review also highlights weaknesses in past studies on the subject and proposes research that would allow future work to be more productive and applicable. Despite our incomplete understanding of the role of cask properties in maturation, the selection of wood or casks on the basis of their effects on flavour is feasible.

Role of oak in the maturation of whisky

The use of oak casks for the maturation of whisky

The whisky industry has traditionally always used oak casks for the maturation of their product and in both the United States and the UK there are now legal requirements for their use. In the USA the bourbon industry is required to store the raw distillate for a year in new, charred oak casks. In Britain, the law demands that Scotch whisky be stored in oak casks for a minimum of 3 years. Legal constraints in both countries prevent the use of flavour additives and discourage the adoption of new production techniques.

The different legal requirements of the UK and the USA reflect very different traditions in the use of casks. The bourbon industry, prevented from reusing old casks, is the main purchaser of new American oak casks. In contrast, the Scotch industry does not generally purchase new casks, depending instead upon the reuse of casks already used for the maturation of other alcoholic beverages. Traditionally, particularly in the nineteenth century, old sherry casks were used, but by far the most common source of oak casks presently used by Scotch producers are old bourbon casks. It is estimated that between 700 000 and 800 000 used bourbon casks are sold every year and although some are used for rum and brandy production, the Scotch whisky industry is a major purchaser. Within the Scotch whisky industry it is estimated that approximately 13 million casks are in use at any time. Oak casks are used for the production of a wide range of alcoholic beverages, including wine, brandy, sherry and rum. These industries use a wider range of oak casks than that used by the whisky industry.

Process of whisky maturation

The effect of maturation on whisky is quite distinct, with the unmatured spirit generally having few of the desirable properties sought in whisky taste and aroma. Therefore the importance of the maturation process should not be underestimated. The following points are known about the maturation of whisky (Nishimura and Matsuyama, 1989):

1 Satisfactory maturation times may vary from 3 to more than 10 years.
2 There is normally a significant flavour difference between matured and unmatured spirits.
3 Volume and strength are lost due to the evaporation of water and alcohol through the porous wood of the casks.
4 Maturation time and the quality of the matured spirit may vary according to the type of whisky, the size, wood type and prior treatment of the cask and the environment in which the whisky is matured.

The mechanisms by which maturation in oak casks occurs are incompletely understood. Research has been carried out to identify compounds that contribute to the flavour or aroma of whisky, referred to as flavour congeners. Correlations between descriptive flavours and chemical analyses of mature whiskies, have identified over 400 flavour congeners (Philp, 1989a). The principle ones are esters, carbonyls, sulphur compounds, lactones, phenols, and nitrogenous bases, including both desirable and undesirable components. In some cases the origin and method of synthesis have been further studied and the involvement of the maturation stage confirmed. Changes in taste or aroma will be due to changes in these flavour congeners. The methods by which this may occur, during the maturation of whisky in oak casks, are listed below (Nishimura and Matsuyama 1989):

1 Direct extraction of wood chemicals.
2 Decomposition of wood macromolecules and extraction of these into the distillate.
3 Reactions between wood components and constituents of the raw distillate.
4 Reactions involving only the wood extractives.
5 Reactions involving only the distillate components.
6 Evaporation of volatile compounds through the cask.

However, as emphasized by Piggott *et al.* (1992) it is the concentration in the 'headspace' (the air space in the cask or container) rather than in the mature spirit that determines the influence on flavour of many volatiles. The concentration of volatiles in the headspace will be influenced by any factors affecting their solubility in the distilate, including the concentrations of involatile compounds.

Canaway (1983) described how the variation of samples from different casks could equal the variation between samples of differing age. Such variation between casks could be due to differences in the raw distillate, the conditions of maturation or the cask wood.

Although both the raw distillate and particularly the conditions of maturation may play an important role in determining the result of maturation, the oak cask in which maturation takes place appears to be of prime importance to the final flavour of whisky (Williams, 1983a). The type of cask can affect both the taste, colour and aroma of whisky. The desired effect of maturation will depend upon the nature of the immature whisky. It may sometimes be desired that the oak wood contribute significantly to the flavour, while for other whiskies, perhaps with an already characteristic taste, the desired effect of maturation may be less. The time taken to reach a satisfactory condition is of financial and practical concern for the manufacturer and varies according to the type of cask used.

Cask and oak types

The source of oak wood used for the construction of casks will normally be one of two general types. Most commonly used is American oak, which is predominantly *Quercus alba*, but may include wood from 10 or more other species of American white oak (Singleton, 1974). The majority of casks used by both the bourbon and Scotch whisky industries will be of this type. Less often used is European oak, consisting of wood from either *Quercus robur* or *Quercus petraea*. Spanish sherry casks may be manufactured from either American or European oak, and it is possible that a single cask may include both types of wood, particularly after repair or reconstruction work.

The division of wood into American and European oak will encapsulate more than simply the botanical species. The two types are associated with different environments and are generally used by very different cooperage industries. It is also important to note that these two simple classifications of cask wood do not form homogeneous groups. There is a long tradition of using different types of French oak for different purposes, as the effect on flavour is thought to vary according to the forest where the oak is grown. However, there is great uncertainty over what determines the different geographic types. Wood from different locations, even if of the same species, may have grown in different climatic or silvicultural regimes. Furthermore the felling and later selection and treatment of wood may vary, with many wood types associated with particular cooperage methods as well as geographic origins. Therefore it is often difficult to discriminate between wood and cask origins. Although most of the oak used derives from the USA or Europe, many other sources of oak have been used for the production of whisky and other alcoholic beverages. Table 1 lists and describes some past and present sources of oak cask wood. Wood other than oak is occasionally used to store alcoholic beverages, although the cask is normally coated on the inside by paraffin or silicone to prevent leakage and the release of unpleasant odours (Knox personal communication). *Robinia pseudoacacia* has been reported as being used for wine casks in Hungary (Lamfalussy, 1953; Molnar *et al.*, 1985), apparently without any coating. Trials in India on the suitability of 12 native timbers to mature whisky found *Terminalia tomentosa* and *Shorea robusta* to be the best substitutes for imported oak. Other woods

Table 1: Sources of oak wood used for maturation of alcoholic beverages

Wood origins	Species reported as being used for cooperage	Main cask uses	Comments
America	*Q. alba* and related white oak species (see Singleton 1974).	Bourbon and subsequently Scotch whisky. Wine and sherry.	Low tannic content but high levels of volatiles.
Western Europe (mostly France)	*Q. robur*, *Q. petraea*.	Wine and brandy.	Varies depending upon precise origins.
Eastern Europe	*Q. robur*, *Q. petraea*, *Q. cerris*.	Wine, brandy, beer.	Present state of oak forestry uncertain—but potentially a major source of cask wood.
Japan and Asia	*Q. dentata*, *Q. crispula*, *Q. mongolica*.	Whisky and brandy.	*Q. crispula* reported to release a sweet taste (Kanazhashi personal communication).
Near East	*Q. mirbeckii* and possibly others.		Oak staves imported from Iran and Turkey during 1940–50s (Williams, 1983b).
South America	Probably *Q. copeyensis* (see Singleton, 1974).	Sherry and whisky casks.	Costa Rican oak reported to have been exported to Spain.

that gave tolerable results included *Quercus dilata* and *W. semecarpifolia* (Anon., 1950).

The cooperage industry

The methods and regulations of cooperage differ between America and Europe, with tighter control generally being found in the supply of American oak. In America the cooperage industry is more automated and operates on a much larger scale than most European cooperages. It accounts for approximately 3 per cent of all the American white oak harvested each year. In the American mid-west 950 000 to 1,200 000 casks per year are made: the majority destined for the bourbon industry (Knox personal communication). In comparison the French cooperage industry, which is by far the largest in Europe, produced around 160 000 casks per year in the early 1990s. However, while new American casks may cost between £50 and £100, new French casks will normally be over £250 each. A variety of construction methods and practices are used, but some of the main steps in cask construction are listed in Table 2.

Table 2: Stages of cask construction in chronological order

1 Selection and felling of suitable timber.
2 Sawing of staves—initial cleaving of wood carried out by some European coopers.
3 Seasoning—air or kiln drying.
4 Precise cutting of staves, bevelling and manufacture of cask heads.
5 Raising of cask—toasting or steaming used to bend staves.
6 Further heat treatments of cask—various heating treatments including the intense charring of bourbon casks.
7 Testing of cask integrity and strength.
8 Use, repair and rejuvenation treatments of cask.

Oak forestry and distribution

Oaks form a major part of the forest flora in both Europe and America. Oak forests frequently exceed over 25 per cent of total forest area in many European countries (France, Greece, UK, Hungary and others) and are often of significant economic value. Their growth, silviculture and exploitation have a long history, particularly in Europe.

Kleinschmit (1993) describes 24 oak species and different hybrid forms existing in Europe. Eight of these are of economic importance, with only the two most important species commonly recognized as suitable for cooperage: *Quercus robur* and *Q. petraea*. These two species are found across most of Europe up to an elevation of 1600 m for *Q. petraea* in the French Alps, with a high degree of range overlap. The two species hybridize, but there is still much controversy over the frequency and importance of hybrids. A similar situation is found among the species of white oaks of north America, where Burger (1975) and others have described the problems of ascribing strict biological species to the various types of oak.

Selection of cask and wood types

The most important factors governing the choice and use of oak casks are practical concerns, such as the ease of supply and economics of use. Therefore, although their previous use is often claimed to contribute to the taste of the mature whisky, the purchase of used casks is due primarily to their low cost. Economic constraints demand the reuse of casks within the industry, despite casks decreasing in viability with each use.

Singleton (1974) claimed that whisky producers have nearly always displayed some preference for the source of oak casks used, but the preferred wood has often been identified simply by the port of importation. Despite widely made claims and traditions that different types of oak will influence maturation in varying ways, at present the flavour effect of oak does not play a major role in the selection of casks. However, the producers of other alcoholic products, particularly wine and brandy, show more discrimination between different types of cask. This is reflected in the much higher cost that French coopers are able to demand for their products, and it is interesting to observe that despite their own sizeable cooperage industry, the USA purchases around half of all French cask exports (Knox personal communication).

Wood properties affecting the maturation of whisky

Wood chemistry

The effects that wood-derived compounds may have on whisky maturation have already been outlined. Flavourful whisky is thought generally to contain high levels of wood-derived compounds. Therefore particular focus has been placed on the role of oak extractives as either flavour congeners in themselves, or involved in their formation or breakdown.

Extractives are compounds found in oak wood that are soluble in either water or organic solvents. There is no precise definition, with the composition dependent upon the solvent used, method and conditions of extraction. The synthesis of most extractive material is associated with the formation of heartwood, and is thought to be controlled by both genetic and environmental factors (Sjoström, 1981). Those compounds believed to be of importance in the maturation of whisky include the following groups.

Tannins and their derivatives The most important group of phenolic compounds are the tannins, being a loosely defined group of water soluble plant polyphenols (Haslam, 1981). The main characteristic responsible for their biological activity, including their astringency (Hagerman and Butler, 1991), is their ability to bind proteins.

The tannins may be further divided into two separate groups. The principal group in oak is the hydrolysable tannins, including gallotannins and ellagitannins, which are described by Hagerman and Butler (1991) as having a polyol (normally D-glucose) as the basic structural unit, of which the hydroxyl groups have been esterified by gallic acid or hexahydroxydiphenic acid (HHDP acid). These tannins are easily hydrolysed either enzymically or in acid or base conditions, to form free gallic or HHDP acid, the latter spontaneously lactonizing to give ellagic acid. Their biosynthesis probably occurs at the transition zone boundary, during the transformation of sapwood to heartwood (Hillis, 1987), with their precursors thought to derive from the shikimic acid pathway (Haslam, 1981; Gross, 1992; Haslam, 1992). The ellagitannins have been found to make up to 10 per cent of heartwood dry weight (Scalbert *et al.*, 1988a). The most common ellagitannins in oak have been identified as vescalagin and castalagin (Mayer *et al.*, 1967), with eight water soluble ellagitannins being characterized by Herve du Penhoat *et al.* (1991a and b). The second category are the non-hydrolysable tannins (condensed tannins or proanthocyanidins). These are oligomeric or polymers of flavonoid units, linked by carbon–carbon bonds, that are not susceptible to hydrolysis (Hagerman and Butler, 1991). In oak heartwood they are found in much lower concentrations than hydrolysable tannins (Scalbert *et al.* 1988a and 1988b).

The solubility of tannins may vary, according to their type, size and various binding reactions with other compounds. Solubility will generally decline with increasing molecular weight. Therefore polymerization is likely to lead to a decrease in the level of soluble tannins. This effect of polymerization has been widely reported for condensed tannins (Hagerman and Butler, 1991), but fewer studies have examined hydrolysable tannins. However, Peng *et al.* (1991) studying tannins in the wood of *Castanea sativa* and *Quercus petraea* concluded that insolubilization of tannins in heartwood probably results from their slow oxidation, leading to polymerization or copolymerization of both condensed and hydrolysable tannins with cell wall components. Such oxidation reactions could be enzymic (involving peroxidases), but in heartwood are more likely to be non-enzymatic. The degree of polymerization would be dependent upon how readily tannins oxidize, with condensed tannins considered more vulnerable to such reactions. Oxidation and polymerization of tannins will considerably modify their astringency and toxicity (Peng *et al.*, 1991). The lower solubility of tannins, due to polymerization, is thought to cause the loss of astringency as fruit ripens (Hagerman and Butler, 1991) and also to explain the reduction in central heartwood durability and resistance to fungal attack (Hart and Hillis, 1972; Peng *et al.* 1991; Scalbert 1992b). Partially oxidized and polymerized ellagitannins may also be responsible for a large part of the heartwood colour (Haluk *et al.*, 1991; Charrier, 1992).

Numerous studies have described tannins in whisky as arising through direct extraction from the wood, the concentration in maturing whisky increasing rapidly in the first 6 months, after which the rate of increase declines (Baldwin *et al.*, 1967; Reazin *et al.*, 1976; Baldwin and Andreasen, 1974). However, the role of tannins in the flavour of whisky and other alcoholic beverages, has not been well established despite the taste often being described in terms of a tannin character. Studies have been further confused by imprecise measurement and use of the term tannins. Most early studies used solely the Folin Denis or Folin Ciocalteau methods of estimating 'total tannins', or more accurately 'total phenolics', which measured both tannins and non-tannin phenolics. Puech *et al.* (1990) showed that the Folin Denis measure of total phenolics gave a good correlation with the tannin content of wood extractive but not for the amounts found in maturing spirits, due to the higher levels of ellagic acid and lignin derived phenolics. Many recent studies report very low levels or no ellagitannins occurring in many spirits (Puech *et al.*, 1990; Ford and Done, 1991; Puech and Moutounet, 1992). Viriot *et al.* (1993) describe the level of ellagitannins in maturing brandies as increasing over the first 5 years, but then subsequently decreasing with further ageing, probably due to chemical degradation through hydrolysis. In a comparison between wine and an alcohol–water solution, both of which had been stored in oak casks for 12 months, Chattonet *et al.* (1989) found that the wine contained lower levels of ellagitannins. This is probably due to reactions with wine constituents, involving binding with proteins or oxidation reactions, whether they be direct, coupled or after hydrolysis. Tannins or their products may be important to the maturation process as oxidative catalysts, or in the removal of sulphides (Chatonnet *et al.*, 1991). The hydrolysable products of tannins, such as gallic and ellagic acids, have been found in many spirits (Jindra and Gallander, 1987; Wilker and Gallander, 1988; Puech and Moutounet), suggesting the breakdown of the hydrolysable tannins. Sefton (1991), describing results of Somers (1990), claimed that in regards to wine maturation the sensory role of oak was not related to total phenolics, while the role of the involatile tannins was unknown. Viriot *et al.* (unpublished) were of the opinion that ellagitannins do not play an important role in the maturation of cognac or other spirits, while Herve du Penhoat *et al.* (199b) thought that tannins contribute indirectly to the taste of brandies, through their complexing or reducing properties. Therefore, despite their abundance in the extract of oak wood, the role of tannins in the flavour of whisky remains uncertain.

A variety of other phenolic compounds are also found in the extract of oak wood. The fluorescent cumarin compound scopoletin is used as an indicator of spirits having been matured in wooden casks (Puech and Moutounet, 1988). A wide range of volatile phenolic compounds, particularly aromatic aldehydes and acids, derived from lignin are also thought important and these are discussed below.

Lignin degradation products A number of compounds found in mature whisky derive from oak lignin. Mechanisms for their formation have been proposed by many authors (Baldwin *et al.*, 1967; Puech *et al.*, 1977; Reazin, 1981; Nishimura *et al.*, 1983; Conner *et al.*, 1989; Nishimura and Matsuyama, 1989; Sarni *et al.*, 1990) and the following pathways have been verified (Nishimura and Matsuyama, 1989) for the origin of lignin degradation products in matured distillate.

1 Degradation of lignin to aromatics due to toasting or charring of casks.
2 Extraction of monomeric compounds and of lignin from the wood.
3 Formation of aromatics by ethanolysis of lignin.
4 Further conversion of compounds in the spirit.

Ethanolysis involves the reaction of the distillate ethanol with lignin, to produce an alcohol-soluble form of lignin. As the solubulization involves the splitting of alkyl aryl ether covalent linkages it is a slow process likely to occur throughout the ageing process (Viriot *et al.*, 1993). Puech and Sarni (1990) outlined three stages in the delignification process:

1 Degradation of cell walls, with lignin polysaccharide bonds breaking and lignin depoly-

merization to dissolvable smaller molecules.
2 Inactivation or repolymerization of small mass molecules, possibly with recondensation on fibres.
3 Subsequent hydrolysis of smaller molecules.

They described an easily extracted lignin complex, that amounted to approximately 4 per cent of the total lignin, and observed that higher levels of tannins appeared to increase the rate of delignification. A variety of phenolic compounds may be produced, which may readily oxidize to give various aromatic aldehydes such as vanillin and syringaldehyde, as well as their respective acids (Baldwin *et al.*, 1967). Puech (1981) described how lignin underwent intense oxidation when in contact with spirits and oxygen, forming aromatic aldehydes. Such reactions may also occur by means of heating or charring the wood.

The role of lignin-derived products on flavour is uncertain, with none of the many studies of them providing conclusive evidence of an effect on flavour. Indeed, their levels in matured distillate are often lower than their individual flavour thresholds (the concentration at which they produce a detectable flavour). However, Maga (1985) found that compounds were synergistic, with a mixture of seven congeners giving a very low mutual flavour threshold of about 2 p.p.m. in 40 per cent alcohol. Therefore, despite individually low concentrations, they may none the less influence flavour. Lignin degradation products also highlight the difficulty in defining the extractive content of wood, with many compounds being formed indirectly through subsequent reactions.

Oak lactones Masuda and Nishimura (1971) identified the two γ-lactone isomers (*cis* and *trans*) as being major components of the volatile fraction of oak wood extractives. These lactones, the so called oak or whisky lactones, derive solely from oak and may be formed from the oxidation of lipids (Maga, 1989b). Tsukasa (1988) reviewed the chemistry of oak lactones, examining a number of different synthesis pathways. These lactones are known to increase in concentration in the distillate during maturation in oak casks, reaching concentrations of up to 10 p.p.m. (Otsuka *et al.*, 1974). Otsuka *et al.* (1974) found a direct correlation between oak lactone concentration and assessed quality scores of different whiskies. Studies on their concentration in red wine (Chatonnet, 1991) suggest they are beneficial to flavour in low concentrations but detrimental in excess, having an aroma of new oak and coconut. Reazin (1981) claimed that their flavour was modified by the presence of furfural. The precise role of oak lactones in whisky flavour is unknown, although relatively high levels in mature whisky are considered desirable.

Acids Measurements of volatile and fixed acids have found that both may increase during maturation (Reazin, 1983). The amount of acetic acid has been found to increase dramatically during maturation (Franco and Singleton, 1984), with studies by Reazin *et al.* (1976) showing that most derives from wood extractives rather than the distillate ethanol. Nykänen (1984) identified dicarboxylic acids as generating aroma compounds and catalysing reactions forming lactones, esters and other compounds.

Fatty acids and other apolar extractives The apolar fraction of oak extract has received relatively little study, despite it being thought that many compounds are important flavour components. The group includes steroids and triglycerides, as well as palmic, steric and oleic acids.

Carbohydrates The levels of many sugars are found to increase in maturing spirits, normally displaying a hyperbolic increase over maturation time (Reazin, 1983). Many of the reducing sugars probably derive from the breakdown of hemicellulose and hydrolysable tannins (Wilker and Gallander, 1988). Nykänen (1984) found that the most abundant sugars in the maturing spirit were glucose, arabinose, and protoquercitol, while other studies (Charrier, 1992) have found fructose and glucose as the most common sugars in oak extract. The thermal degradation products of cellulose and hemicellulose, such as furan and pyran volatiles, have also been reported frequently in whisky, most recently by Clyne *et al.* (1993). However they are not thought to have a major influence on flavour (Chatonet *et al.*, 1991).

Nitrogenous compounds Both polyphenoloxidase and peroxidase activity have been found in heartwood of oak (Ebermann and Stich, 1992). Amino acids and other nitrogenous compounds, such as pyrazines and pyridines (Maga, 1985) have been detected in charred oak extract, with concentrations of 17 and 2 mg respectively per 100 g dry weight of wood. Although it has not certain that they influence flavour, such compounds are known to have very low flavour thresholds.

Terpenes This group of volatile compounds has received relatively little attention, despite being important flavour and colour compounds in spices, perfumes and other aromatic products. Studies by Sefton *et al.* (1990a), Nabeta *et al.* (1986) and Nishimura *et al.* (1983) have identified monoterpenes, sesquiterpenes and various norisoprenoids among the constituents of oak wood. Sefton (1991) describes the norisoprenoid group as the most diverse and identifies, in oak extract, precursors of compounds patented as flavour additives.

Other compounds A range of additional compounds found in the extractive content of oak may be of relevance to whisky maturation, but have yet to receive attention. Those of importance include both volatile and non-volatile compounds. Over 200 volatile components of cask wood have been identified (Sefton *et al.*, 1990a), and many others may be present. Non-volatiles, such as tannins, may influence flavour by direct or indirect effects, such as affecting the solubility of volatile compounds (Piggott *et al.*, 1992). As already indicated, while whisky may obtain some flavour congeners by direct extraction, many others, particularly aromatic aldehydes and their related acids, will derive from both extraction and further reaction of compounds from the cask.

Wood anatomy

Effects of wood anatomy The use of oak wood for the manufacture of casks is primarily due to its ability to contain liquids with little leakage. The wooden cask must also meet other cooperage criteria, such as suitable strength and flexibility. Anatomical features could influence the maturation process through two possible mechanisms. Firstly the location of extractive deposits and any factors that influence the permeability of wood, are likely to affect the availability of wood extractives to the maturing distillate. Secondly, the cask wood will influence the maturation conditions and environment. For example, any features that influence the movement of gases through the wood will affect both the rate of evaporation of the distillate and the availability of oxygen. As well as influencing whisky maturation directly, anatomical features may correlate with other properties that affect maturation. If chemical requirements can be shown to correlate with physical properties the selection of wood for flavour effects could be easier.

Anatomy and properties of oak wood Oak wood is ring porous. The early wood is laid down at the start of the growing season and consists mostly of large vessels. The late wood has a greater proportion of fibres and only small vessels are present. The rate of growth determines the size of annual rings and the proportion of late to early wood increases with ring width. Sapwood, containing living parenchyma cells and frequently starch, eventually transform into heartwood. This transformation involves cell death, the removal of starch and laying down of extractives. Structurally the woods are very similar, despite the lower permeability, greater durability and darker colour of heartwood. The heartwood periphery may be undulatory, and does not normally correspond with a specific growth ring.

Tyloses are occluding structures found in vessels which develop through pit apertures from adjacent parenchyma cells. The formation of tyloses is generally associated with the conversion of sapwood into heartwood. However they also form in the sapwood of felled timber during storage to an extent depending upon the conditions and duration of storage (Alexander, 1972; Bolton personal communication). Tyloses may also form as a response to fungal infection or injury. Wheeler and Thomas (1981) report the findings of Williams (1942) who found that in white oak the late wood vessels rarely have tyloses, in contrast to the larger early wood vessels. The precise stimulus for their formation remains uncertain, with exposure to air and changes in the concentration of ethylene among the proposed causes (Hillis, 1987). Tyloses are not found in the wood of all species of oak, with *Q. rubra* being a notable case, making their wood unsuitable for leak-proof cooperage.

The number and structure of wood rays are thought to influence the radial permeability of wood, possibly to both gases and liquids. Rays may consist of either a single (uniserate) or 5–30 (multiserate) rows of cells across a transverse section and be hundreds of cells in height. The size of rays was found by Feuillat (1991) to be inversely correlated with their number.

Permeability Low wood permeability is necessary in order to ensure a tight cask with little leakage. The impermeable nature of oak wood, together with its wide availability, were probably the main reasons for its initial use. Tyloses are generally considered to be the primary cause for heartwood impermeability to both gases and liquids. Lehmann (1988) found that air permeability of *Fagus syvatica* heartwood was closely correlated to tylose formation. Likewise Kuroda *et al.* (1988), in studying twenty hardwood species, found that the presence of tyloses in the wood could be predicted from measuring permeability. The ring porous nature of oak may also be important in restricting its permeability.

The most relevant measure of permeability is the tangential permeability, which corresponds to movement through a stave, from the inside to outside of a cask (see Figure 1). The size, abundance and distribution of vessels, degree of closure by tyloses, the number of wood rays and other anatomical factors are all likely to influence tangential permeability. Due to the ring porous nature of oak, the size of annual rings may also be important, as this will determine the relative abundance of large vessels, which are found only in the early wood. The anatomy of oak wood is very variable, even when the ring size is similar.

However, studies examining the permeability of wood in relation to specific anatomical features have rarely found any strong correlations. Kuroda *et al.* (1988) found that neither vessel radius, nor percentage of vessels by volume nor total number of vessels per cross sectional area, gave good agreement with permeability in ring porous woods such as oak, which have high variation in vessel size. Sato *et al.* (1990) studied the penetration depth of cask staves by malt whisky, which correlated with neither ring widths nor the alignment of the wood relative to the radial direction. The dominant role of tyloses in determining the permeability through wood may explain the failure to detect such correlations. In contrast Maga (1989b) stated that it was the preponderance and spacing of large rays that made it difficult for liquid to pass through oak wood. The importance of rays in liquid or gas movement in wood is uncertain, although the estimated 25 per cent lower tangential permeability compared with radial permeability is thought to be due to rays allowing greater radial flow (Kumar and Kohli, 1988). Wheeler and Thomas (1981) suggested that the permeability of rays depends on their width, with single cell uniserate rays being the most permeable. Other features that are thought to influence permeability include the deposits of extractable material, which may block vessels and cell pits (Wheeler and Thomas, 1981; Kumar and Kohli, 1988).

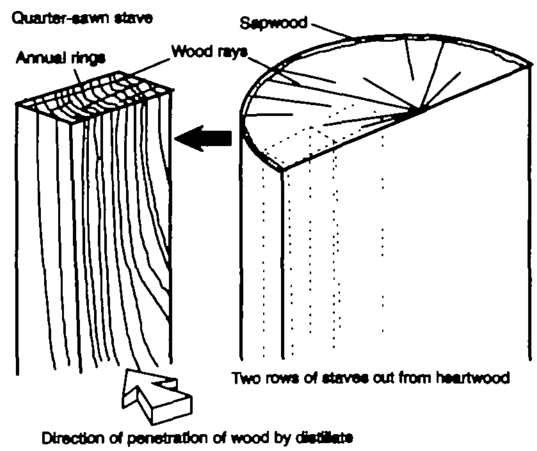

Figure 1. Alignment of stave wood.

The permeability also varies between different liquids. Kiseleva and Zoldners (1986) found that pure water diffused through birch wood four times faster than pure alcohol, when restricted to passing through cell walls, as is likely to be the case when movement through vessels is blocked by tyloses.

Wood density Zhang *et al.* (1993) describe the close correlation of oak wood density with both ring width and cambial age (the number of rings away from the pith). Ring width influences density predominantly by the ratio of early to late wood, with the early wood having a lower density than the late wood. Quickly grown oak, with wide rings, tends to produce wood with a higher density than slowly grown oak. Keller (1987) described the best cask wood as being less dense and therefore more permeable than wood used for most other applications.

Wood colour Klumpers *et al.* (1993) found a tentative correlation between wood colour and extractive content, with heartwood colour being loosely correlated with the tannin content. The darker colour of heartwood is frequently claimed to be due to the laying down of phenolic extractives. The colour of the wood almost certainly correlates with some measure of extractives, as the wood becomes visibly paler after extraction with ethanol or acetone solutions. The majority of studies addressing the colour of oak wood have focused on the problem of discolouration during seasoning of the wood (for example Haluk *et al.* 1988 and 1991). Charrier (1992) found that brown discolouration was associated with a decrease in the levels of ellagitannins, with a corresponding increase in the concentration of ellagic acid and possible coloured degradation products.

Wood grain This empirical term is much used among wood traders and coopers to describe cask wood. The grain of the wood is determined by the visual impression produced by the size of wood elements, particularly vessels. Different types of grain may include fine, coarse, tight and loose. Feuillet *et al.* (1992) describe how the term has varied in its use over history and how it presently most often refers to the ring width and wood texture. This in turn relates to the porosity of the wood and the abundance and distribution of large vessels. Although the term lacks objective precision, none the less its continued use among professionals, particularly those working in French forests, makes it important to understand the properties to which the term relates.

Summary of the role of wood in the maturation process

The properties of the cask wood clearly have a major effect on the maturation process. Of particular importance, and having been the subject of numerous studies, is the amount and composition of the extractive content. Various flavour congeners are thought to derive directly or indirectly from extractive compounds and a number have been identified as being of likely importance. Anatomical characteristics of the wood may also influence the maturation process, particularly the rates of evaporation, oxidation and extraction. Any factors that cause variation of these properties will thereby influence the maturation of whisky.

One cannot define a set of properties, be they chemical or physical, or the cask wood that may be used as criteria in the selection of wood suitable for whisky maturation. However, a number of characteristics are accepted as influencing maturation. One can examine the variation in these characteristics to determine the feasibility of selecting wood for its effects on maturation. The next section examines evidence for variation in relevant properties and likely causes and patterns in this variation.

Factors determining cask properties

Numerous studies have compared the effect of using different types of cask and many have found apparent differences. However it is often found that numerous factors such as the method of cooperage, age and drying of the wood, in addition to the source of oak used, vary between cask types. Therefore although many studies have shown that there is variation in cask properties and effects, the source of this variation has not often been identified. An attempt will be made to discriminate between variation in the properties of the virgin wood and that among used casks.

Variation in oak wood properties

Variation between species Many studies examining variation of the properties influencing maturation fail to identify the species of oak from which the wood derives. Frequently oak wood is referred to as simply American or European. Despite this there is evidence that a range of properties appear to vary between at least some species, although few studies have compared the most relevant species: *Q. alba*, *Q. robur* and *Q. petraea*.

Azizol and Rashid (1981) claimed that there is a common pattern of fluorescent phenolics for most hardwood species and Salagoity-Auguste *et al.* (1986) were able to distinguish between chestnut and oak by the ratio of gallic to ellagic acids. Kishimoto and Kitiamura (1973) used infra-red and ultraviolet absorption spectra of different extracts to classify 13 *Quercus* species into four main groups.

Similarly, Knops and Jensen (1980) found that phenolic variation supported morphological evidence of hybridization in three species of red oaks, while Li and Hsaio (1973) described how the leaf phenolics differentiated between the subgenera of American oaks.

Although some studies have found no flavour differences between American and European casks (for example Aiken and Noble, 1984), there are many others, which have controlled other sources of variation, to conclude that there are distinct differences between American white oak species (such as *Q. alba*) and the European species *Q. robur* and *Q. petraea*. Guymon and Crowell (1970) described clear differences in the effects of the two types and their results are shown in Table 3.

Rous and Alderson (1983) also reported differences between French and American oak casks, each constructed identically by the same cooper. Both the quantitative differences given in Table 4 and qualitative differences in the taste imparted to the wines were found.

Table 3: Composition of brandy matured for 72 months in American or European oak casks

Component	New US standard cask	New French Limousin cask
Tannins	56	102
Esters	72	61
Furfural	4	1.4
Total extract	176	232
Colour	11	20

From Guymon and Crowell (1970).
Umts, mg/100ml; colour, units relative to standard.

Quinn and Singleton (1985) also found levels of ellagitannins extracted from French oak were greater than those from American oak. Variation in the proportions of different ellagitannins was also found. Puech (1984) likewise found that the levels of tannins from American oak were less than European oak, as did Singleton (1974). Puech (1984) found the level of non-tannins was greater in American oak.

Table 4: Gain in phenolics (mg Gallic acid equivalents / l) found in white wine after 13 weeks maturation in either French or American casks

Extractives measure	1st fill French cask	1st fill American cask
Total phenolics	231	56
Non flavonoid phenolics	144	49

From Rous and Alderson (1983).

Nabeta *et al.* (1986) compared the volatile components of heartwood from the four species *Q. robur*, *Q. mongolica*, *Q. dentata* and *Q. serrata*, and found that *Q. robur* had the lowest concentrations of oak lactones, and furfurals. Guymon and Crowell (1972) suggested that American oak contained higher levels of lactones than European oak. Nabeta *et al.* (1986, 1987) reported that *Q. robur* contained five times the amount of the most commonly occurring phenolic, lyoniresinol, than did American oak. In another comparison between the two types, American oak has been found to contain greater concentrations and variety of volatile norisoprenoids, than three types of French oak (Sefton *et al.*, 1990b; Sefton, 1991).

Comparisons between the two European species, *Q. petraea* and *Q. robur*, have given less clear results. Chatonnet (1991) reported that the average levels of lactones and eugenol were in the region of four times greater in *Q. petraea* than *Q. robur*, concluding that the wood of *Q. petraea* was more aromatic. Chatonnet (1991) also indicated that *Q. robur* was richer in phenolic extractables, particularly ellagitannins. However, Feuillet (1991) expressed reservations on the validity of the studies, as they do not adequately differentiate between variation due to species and that due to geographic origins.

A few studies have examined variation in relevant physical properties, such as wood permeability. Comparisons between American and European species claim that American oak has more tyloses and is less permeable (Rickards, 1983). Various comparisons between physical properties of *Q. robur* and *Q. petraea* have also been made, including Keller (1987). There is much overlap of characteristics, and few properties act as a reliable guide to distinguishing between the two species. Indeed, as already discussed, there is still much controversy over the occurrence and abundance of hybrids and intermediate forms.

Variation within species Studies indicate variation in both chemical and physical wood properties within species, but it is unclear how this reflects differences between geographic races, populations and ecotypes. The relative importance of environmental and genetic differences also remains uncertain.

A number of studies have reported geographic differences in flushing time in European (Burger, 1949; Liepe, 1993) and American oaks (Kriebel, 1993). Kleinshmitt (1993) described how wood properties, including density, have been found to relate to flushing time (Jevlev, 1972a,b; Nikolov *et al.*, 1981). Studies on Douglas fir describe how wood permeability declines significantly with altitude of growth (Polge, 1973), and it is possible that oak may be similarly affected. McDougal and Parks (1984) showed that leaf phenols of *Q. rubra* varied between sites at 75 and 1140 m elevation. Variation in growth rates have also frequently been reported from oak provenance trials (for example Barzdajn, 1993; Gracan, 1993; Jensen, 1993). These geographic differences are frequently found to be greater than corresponding species differences. Kalinkov and Shipchanov (1976) in comparing anatomical properties, such as vessels and fibre abundance of *Q. petraea* and three other species of oak, found the greatest variation to occur between moisture loving and drought hardy forms of *Q. petraea*. However, many authors report greater ecotypic variation between local populations, than geographic or clinical variation. Baranski (1975) found variation in several properties of *Q. alba* to be greatest within regional populations than between them and no evidence of altitudinal ecotypes of this species. Kriebel (1993) also emphasized ecotypic variation among American oaks, while Kleinshmit (1993) and Jensen (1993) stress a similar pattern of variation in European oak populations.

A number of studies indicate the importance of various environmental factors that may account for this local variation. Feuillet (1991) summarized some of the conclusions of Henry (1886, 1887, 1892 and 1896) which were that, all other factors being equal, oak wood was richer in tannins when the trees grew on calcareous soils or were isolated and more exposed to light. This is in accord with Mallet (1946) who claimed that oaks in full sunlight gave rise to a better quality armagnac. Keller (1987) examined morphological variation in French oak and identified the rate of growth as the main cause of differences between oak types. He also described some oaks as producing low density wood despite rapid growth, with few fibres being laid down in the late wood: a trait he considered to be genetically determined. Nepveu (1993) also emphasizes the genetic control of wood properties and claims that the large phenotypic variation cannot be explained purely by variation in growth rates or environmental factors. Studies on the heritability of various oak wood properties have been summarized in Savill and Kanowski (1993). Recent work (Mosedale *et al.*, in preparation) suggests that heartwood tannin content is under strong genetic control.

Many authors have described very high levels of gene diversity within oak species and populations, with high individual tree variation (Olson, 1975; Kremer and Petit, 1993; Muller-Stark *et al.*, 1993). However, Zanetto *et al.* (1993) reported that the pattern of genetic variation itself varied between regions of Europe. Kleinschmit (1993) noted that the long history of human exploitation and planting has further encouraged diversity, in addition to the life history characteristics of oaks that encourage outbreeding of populations.

Studies focusing on possible flavour-influencing properties have frequently given ambiguous results. Despite claims that tannin content is more variable between different provenances than different species and the tradition of identifying French oak according to its geographic origins, recent comparisons between geographic types of cooperage oak have produced conflicting results. Puech (1984) compared extractives of different types of European oak with American oak (see Table 5) but, due to low replication, the study fails to show any clear evidence of variation between geographic types of European oak, although it does demonstrate the high level of variation between individual trees. Miller *et al.* (1992), in comparing wood of *Q. alba* and *Q. robur* from two American forests, found significant differences between the sites in total phenols and some phenolic acids. However, site differences were not as important as species differences and only two trees, of each species, were selected from each site.

Puech (1984) found lower levels of coniferaldehyde and sinapaldehyde in French oak than American, Russian, or Bulgarian oaks, while greater concentrations of lactones have been reported in Russian brandy than French brandies. It might be expected that lignin degradation products will vary little between oak origins or species, due to the common source from which they derive. However, Puech and Sarni (1990) suggested that the fraction of easily extractable lignin, from which these compounds derive, could vary between different provenances.

French oak from the Limousin region and from the forests of central France (frequently referred to as Tronçais or Allier oak) are the two types most frequently claimed to cause different flavours. More than referring to oak from a particular geographic region, these two types typify opposing characteristics with regard to the species, silviculture, location, and age of exploitation of the forests (Feuillat 1991; Pontallier 1991; Remy 1991; Giraud personal communication). Feuillat (1991) reviewed studies on the properties of these two types and his conclusions are summarized in Table 6.

In general, there is evidence for variation between both geographic regions and local populations in wood properties affecting flavour. However there is much uncertainty over the pattern, degree and causes of this variation.

Variation within trees Variation within trees is frequently ignored and yet it is known that this may be significant for many properties. Many authors (for example Peng *et al.*, 1991) describe how the concentrations of soluble and insoluble ellagitannins respectively decreased and increased, from the periphery of the heartwood towards the centre. The results corroborate earlier studies on the distribution of tannins by Schultz (1959) and Henry (1886, 1887, 1892, 1896). It is thought that the tannins slowly oxidize and polymerize as the heartwood ages, thereby becoming less soluble. Similar reactions may also affect the levels of other extractives, but few studies have been undertaken, beyond simple comparisons of sapwood and heartwood, such as Maga (1989a) who reported levels of oak lactone to be higher in heartwood.

The speed of growth varies with age, and therefore properties that are influenced by ring width may vary likewise. The degree of variation will depend upon the regularity of growth and can vary not only between growth rings,

Table 5: Total and percentage extractable tannins of different oak origins

Type of oak	Tannin content (mg/g wood)	Means	Extractable tannins (percentage of total)	Means
Troncais	84, 96, 135	105	62.4, 65.2, 63.7	64
Limousin	73, 89, 154	105	41, 66.1, 66.5	58
Gascony	80, 82, 105, 111, 120, 150, 153	114	47.4, 48.7, 54, 57.7, 58.3, 60.2, 60.7	55
Bulgarian	79		49.9	
Russian	105		55.5	
American	35		35	

Derived from Puech (1984).

Table 6: Summary of studies on the properties of Limousin and Allier oak

Characteristic	Limousin oak	Tronçais or Allier oak
Climate	Moist, high rainfall.	Drier climate & environment.
Silviculture	Both coppice and high forest systems. Greater thinning than Allier.	High forest—with a greater tree density than Limousin.
Age of trees used	90–120 years	200–250 years
Species	Mostly *Quercus robur*	Mostly *Quercus petraea.*
Physical properties		
Grain	Coarse/loose	Fine/tight
Ring width	2.5–5 mm	1–2 mm
No. of lines of large vessels	2+	1–2
Size of large vessels	Larger	Smaller
% Open vessels	Greater (fewer tyloses)	Fewer (more tyloses)
Ray no. & size	Equal	Equal
% fibres	Equal or greater	Equal or fewer
Wood colour	Yellow	Pink/rose
Tightness of cask	Less: possibly due to fewer tyloses in large vessels.	Greater
Permeability	Higher	Lower
Extractables	Greater exchange with spirits.	Less
Chemical properties		
Total extractives	140 mg/g wood	90 mg/g wood
Polyphenols (D280)	30	22
% Tannins	10	6
Ellagitannins	15.5 mg/g wood	7.8 mg/g wood
Colour of extract	Yellow	Pink
Lactones	17 g/g wood	77 g/g wood
Phenolics	Greater	Less.
Aromatics	Less	Greater
Aroma	Less expressive, and not as complex.	Richer, more intense.
Taste	Astringent	Pleasant, complete
Notes on use	Used for cognac & eaux de vie, as tannins released rapidly.	Used for wines, as resulting flavour is released more slowly, and therefore more easily controlled.

Derived from Feuillat (1991).

but also within a ring if radial growth is assymetrical. In addition to noting that any direct relationship between ring width and phenolics will be complicated by the phenolics being laid down in the wood 10–15 years after the growth ring is formed, Singleton (1974) described how greater amounts of phenolics were extracted from the sawdust of early wood than late wood. These results suggest that one would expect slowly grown oak to contain greater concentrations of tannins due to the higher proportion of early wood. One might also expect slow grown oak to be more permeable (and therefore release more extractives) as the density of early wood is much lower, containing as it does the large vessels. Singleton suggested the speed of growth may explain the lower extraction and less leakage of casks made from American oak, as they have a reputation of being grown faster than European oaks. However variation of many wood properties influenced by ring width, such as density and shrinkage, have frequently been found to vary more between individual trees, than within. Zhang *et al.* (1993) describe how the relationship between wood density and ring width can vary between different trees.

Summary of variation in oak wood

There is evidence for variation in properties affecting whisky maturation both between and within species of oak. Less certain are the causes of such variation, and also the significance of the different levels of variation, be they between species, provenances or individual trees. However, there is sufficient evidence to conclude that the species of American oak vary in chemical and possibly anatomical properties from the European species, *Q. robur* and *Q. petraea*.

Feuillat (1991) in studying a number of properties of French oak concluded that any geographic effect on both physical and chemical factors was due primarily to differences between species. Therefore *Q. robur* from the St Palais region was more similar to *Q. robur* from the Limousin than it was to *Q. petraea* from the same region. The considerable differences often attributed to Limousin compared to Allier oak could be primarily due to *Q. robur* being most abundant in the Limousin, while *Q. petraea* is dominant in the Allier region. However this in turn is not a very satisfactory conclusion given the difficulty of species identification and the known hybridization that occurs between the two main European species. It is reasonable to conclude that provenance differences exist, but the degree to which these are genetically or environmentally determined remains uncertain. Furthermore it should be remembered that nearly all studies highlight the large degree of variation between trees within the same population.

Variation in cask wood properties

Among the various factors that may alter the extractive content of wood between its felling and the use of casks, only those considered to be most important are discussed here. Some of the main effects are summarized in Table 7.

Selection of wood

The selection of wood for cooperage imposes a number of restrictions on the wood used for whisky maturation. Generally oaks with a diameter at breast height of not less than 35 cm

Table 7: Summary of the effects of cooperage treatments on wood properties

	Treatment effects			
Treatment	Tannins	Oak lactones	Other extractives	Anatomical properties
Seasoning	Decrease in soluble tannins during wood ageing widely reported.	Lactones reported to increase during wood aging.	Conditions of drying reported to influence volatile constituents.	Decrease in moisture content to approximately 14% of dry weight.
Toasting	Decreases levels of ellagitannins through oxidation or hydrolysis reactions.	Heating may increase synthesis, but there will be loss through volatilizing from the wood surface	Degradation of lignin leads to the formation of lignin derived compounds available for extraction. Levels of volatiles reported to increase.	At high temperatures physical degradation of the wood may occur. Effects will vary through the depth of the stave.
Charring	Decreases ellagitannins through the same reactions as occur in toasting.	Conflicting reports about the effect on lactones.	Large amounts of furan and lignin derived compounds formed. Charred layer may also remove undesirable sulphur compounds.	Break up of wood structure near surface may allow easier penetration of new wood and extraction of compounds.

are used (Keller, 1987). Only the oak heartwood is used and this is normally explained by the formation of tyloses and the laying down of extractive material, which reduce wood permeability and ensure a tight container. High quality oak wood is required, free of defects such as knots, frost or fungal damage. Trees with spiral grain are also considered less suitable, as the vessels may not run longitudinally along the resulting staves. Coopers may display other preferences, so as to reduce splitting or checking in the wood. However these vary so widely that Alexander (1972) found no common guidelines among various American coopers. By selecting only high quality oaks with suitable characteristics, limitations are placed on the range and type of variation in properties influencing flavour.

Manufacture of staves and cask

Claims are frequently made, particularly by European coopers, that the initial cleaving of the wood is preferable to sawing. Reasons for this include that sawn wood gives rise to 'strange aromas' (Castelli and Peynaud, 1990) or that cleaving staves helps prevent leakage from the finished cask by ensuring that the vessels are correctly aligned along the wood (Edlin, 1973). While this latter claim does not seem unreasonable, the American coopers are able to produce tight casks without cleaving. Sawing has the added benefit of producing less waste (Williams, 1983b) and is more easily mechanized. Furthermore the fact that Portugese coopers produce casks from sawn staves of Limousin oak, discounts the possibility that cleaving is a requirement for *Q. robur*, if not *Q. alba*.

Quarter-sawing staves, so that rays run horizontally across and vessels longitudinally along the stave (see Figure 1), appears a common practice among coopers. This is often claimed to reduce leakage, by restricting movement along the wood rays to going across, rather than through the stave. However, Howard (unpublished) reported that quarter-sawing also decreased the amount of warping and the effect of any splitting parallel to the rays.

Finally it should be noted that a finished 190 litre cask will normally contain in the region of 31 pieces of wood, including both staves and headings. These are all likely to derive from different trees, or even provenances, and therefore the variation in cask effect may be less than if each cask derived from an individual tree.

Seasoning

The method of seasoning wood is often said to affect both the structural integrity of the wood and the flavour imparted by the cask. Glaetzer (1991) claimed it was the most critical factor in oak selection for wine maturation. The traditional, European method of seasoning is by air-drying, taking approximately 3 years to reduce the moisture content of staves to the required level of approximately 15 per cent (Remy, 1991; Pontallier, 1992; Giraud personal communication). The time required varies according to the climate and has been reported as taking only 9 months in the USA. Air-drying was adopted because of the problems associated with kiln-drying oak wood. Casks made from wood that has been seasoned too quickly are liable to be brittle or to develop open joints (Panshin *et al.*, 1962). Better control of kiln-drying now allows oak to be seasoned successfully in this way, and although structural defects and discolouration can still present problems (Charrier, 1992), the speed of kiln-drying has the major advantage of allowing the cooper to meet demand more easily. The process normally involves leaving the staves in a ventilated drying room at 40–60°C (Pontallier, 1992).

Many wine producers still express a preference for air-dried wood, it being frequently claimed that the flavour produced is more subtle and complex than that from kiln-dried oak. Studies comparing the results of kiln and air-dried wood give conflicting results. While some have found no significant differences (Wilker and Gallander, 1989), other studies claim to have done so. Pontallier *et al.* (1982) found that air dried Allier oak resulted in lower levels of phenolic acids being extracted by wine than occurred in kiln dried oak. Sensory comparisons of oak-aged wines also found that air dried wood gave a stronger 'vanillin wood' character than the 'green and dusty' kiln dried casks.

Many recent studies have found the effects on flavour of drying depend on the seasoning method and conditions. Sefton *et al.* (1990b) found that the conditions of air-drying were important, reporting that levels of oak lactone decreased in oak seasoned in France, but were unchanged when the same type of oak was seasoned in Australia. Differences in other volatile compounds also appeared to depend on seasoning conditions. They concluded that although the reasons for such variation were unknown, the local climatic conditions such as humidity and temperature may have a major effect. A number of studies appear to indicate that the age of the wood after felling is the most important seasoning factor influencing extractive content. According to Maga (1989a) significant loss or degradation of particularly unsaturated compounds, fats and fatty acids occurs during ageing. He also reported that the concentration of oak lactone increased five fold during 6 years of seasoning American oak under cover. Sefton (1991) described how the levels of vanillin in oak seasoned for 2 years were twice those found in green oak, while the amount of eugenol decreased. A decline in tannins during wood storage and air drying was reported as early as the last century (Jolyet, 1892). Theories proposed to explain this phenomenon include enzyme activity causing condensation and polymerization of tannins, or that when exposed during air drying rainwater leaches out soluble tannins (Wilker and Gallander, 1989). Pontallier (1992) suggested that hydrolysis and oxidation reactions, provoked by the action of enzymes in the wood and those secreted by micro-organisms which develop during air-drying, causes condensation and polymerization of tannins. However, Peng *et al.* (1991) suggest a decrease in soluble tannin content is most likely due to non-enzymatic oxidation reactions in the heartwood, causing the insolubilization of tannins. These changes will occur to a lesser extent when kiln-drying as the process takes less time. It would explain the lack of significant differences found between air and kiln-dried wood by Wilker and Gallander (1989), as the kiln dried oak was stored for an additional 2 years, while the air-dried samples were seasoned. These studies suggest air-drying causes a decrease in the soluble tannins, which although perhaps beneficial for many wines, might be considered undesirable for maturing spirits such as whisky.

In summary it appears that the age of wood after felling may be of great significance, with the conditions of seasoning also playing a role in determining flavour effects. Furthermore, different oak wood appears to respond to seasoning in different ways (Sefton, 1991). This is in agreement with Skurriken *et al.* (1970) who, on evaluating the quality of Bulgarian and Russian oak wood for brandy ageing, reported that both the origin and seasoning influenced the brandy quality. The wood judged to be most suitable for brandy maturation had been stored for 17 years before being made into barrels.

Charring and toasting of casks

Charring is carried out on new casks by American coopers for the bourbon whisky industry and also as a means of rejuvenating used whisky casks. Toasting occurs during the heating process (bousinage) of bending the staves, but is frequently continued beyond the level required for this purpose. John (1991) claimed that slow constant heating was preferred for the manufacture of barrels for wine storage, but the duration of heating may range in time from 20 to 60 min, with many coopers offering their customers varying degrees of toasting. However the toasting of casks is usually an entirely empirical process, and therefore there is no objective ranking of toast between different coopers.

The charring of casks dramatically affects the volatile composition of the oak wood, and increases the levels of many cask extractives. The flavour derived from charring has long been thought favourable in whisky maturation, as shown by recent studies (Clyne *et al.*, 1993). Many studies (Baldwin *et al.*, 1967; Singleton, 1974; Marsal and Sarre, 1987; Sarni *et al.*, 1990; Clyne *et al.*, 1993) have found that concentrations of furan and lignin degradation products are higher in spirits matured in charred casks. Maga (1989a) proposed that thermal lipid oxidation would give rise to lactones. According to work by Ford and Done (1991) the charring of wood also eliminates, or greatly reduces, ellagitannins from the extractive content. Another reported effect (Paterson and Piggott, 1989) is that charring forms a layer of active carbon that may remove undesirable flavour congeners.

Rejuvenating used casks by re-charring increases the amount of colour, solids, fixed acids, tannins and aromatic aldehydes that can be extracted, increasing the viability of the cask. However, the same levels as those found in a new cask will never be reached and viability will once again decline with reuse.

Similar effects are reported by studies on toasting, although some apparently conflicting results have been obtained. Chatonnet *et al.* (1990) found that while toasting increases the levels of lignin degradation products and furan products, it decreases the levels of polyphenols and lactones. Marsal and Sarre (1987) in addition to reporting an increase in the furan products, furfural and 5-hydroxymethyl furfural, also noted decreased levels of oak lactones extracted from toasted wood. Maarse and van der Berg (1990) described how pre-treating wood by heating decreases tannin concentrations, giving a less bitter taste.

The effect of varying degrees of charring and toasting has also been studied. Nishimura *et al.* (1983) found that although the amount of aromatic aldehydes extracted from heat treated wood increased with temperature, the amounts decreased when charring of the wood had occurred; although levels were still higher than those from untreated wood. Chatonnet *et al.* (1989) also found that the maximum levels of phenolic aldehydes occurred at medium to high levels of toasting, with very high toasting decreasing the aromatic impact of vanillin, presumably due to the conversion of aldehydes to their respective acids. However he found that the effect of toasting varied between different wood types, with greater toasting of Allier oak decreasing the levels of total aromatics, compared to Limousin oak where these increased while the tannic flavour of the wood declined. Sarni *et al.* (1990) found that increased heating led to a decrease in the levels of ellagitannins and a corresponding increase in ellagic acid. Their results, describing the effects on lignin of increasing temperature, suggested the progressive formation of different degradation products. From a temperature of 120°C, initially aldehydes would appear followed by an increasing abundance of acids at temperatures of over 165°C. Thermolysis would occur at higher temperatures according to the monomer units being considered, with firstly guaiacyls and later syringyls, degrading to form guaiacol, dimethoxyphenols, cresols and other phenol type compounds characteristic of burned wood. They noted that spirits such as whisky, matured in casks subjected to direct charring, contained higher levels of syringyl compared to guaicyl units, as well as containing typical pyrolysis products. Levels of the lignan lyoniresinol appeared unaffected by heating.

When considering the impact of either toasting or charring, two distinct effects are likely. First there will be the effect of heat on the wood chemistry, the degree of which will vary throughout the depth of the treated staves, thereby ensuring a complex range of effects upon the cask. Second, at high toasting levels and during charring, there will be physical degradation of the wood, which may allow increased penetration by the distillate.

In summary, the heating of wood appears to increase levels of lignin degradation products, aldehydes, acids, furan products and many volatiles; the effect of different temperatures varying according to the molecular structure. At high levels of toasting, or charring, thermolysis products will develop. Phenolics are likely to undergo oxidation or possibly hydrolysis reactions, decreasing the amounts of soluble tannins, particularly ellagitannins. The effect on oak lactone appears to be uncertain, for while some studies suggest charring of a new cask increases levels, studies on toasting and rejuvenating casks by charring suggest that the treatment results in little increase. The physical degradation of the wood by charring may be the cause of increased extraction of many compounds, particularly when charring is used to rejuvenate old casks. The layer of charcoal may also remove undesirable compounds from the maturing distillate.

Other cooperage effects

A range of treatments may influence wood properties, in varying ways. Methods other than toasting barrels are used for the bending of cask staves, particularly in America where the wood is pre-heated by steaming for 10–15 min (John, 1991). This is likely to influence the cask properties in a different way from toasting. Immersing the staves in hot water is practised in Hungary, resulting, it is claimed, in less harsh tannins and a more subtle pickup of flavour by the wine (Degaris, 1991). It is probable that the treatment would cause the removal of some water soluble extractives, particularly tannins. Similar effects may be caused by the practice of testing the tightness of the finished cask by partially filling under pressure with hot water or by sterilizing treatments which are often carried out by cask users.

Previous use and rejuvenation of casks

It is accepted that the levels of cask extractives decline with reuse and that the quality and speed of maturation is strongly influenced by this decline (Puech and Visockis, 1986). Decline in cask viability has led to the adoption of various methods of rejuvenation. The most common one, involving the re-charring of casks, has already been discussed. Another frequently used method is scraping the inside of casks, removing the inner surface, possibly in conjunction with recharring. Examination, by electron microscopy, of the inner and outer surfaces of an oak stave used to mature Armagnac for the previous 80 years (Puech, 1984), indicated that the process of maturation did not appear to alter the structure of the wood. Furthermore the levels of extractives had only been decreased in the wood to a depth of approximately 9 mm. This depth is typical of the distance travelled by the spirit front, easily observable in used staves. It confirms the effectiveness of scraping as a method of rejuvenation, as it would expose new unextracted wood to the distillate. However the treatment may weaken the cask and cannot be repeated frequently.

In apparent contradiction to the detrimental effects of reuse, the previous storage of some alcoholic beverages, such as sherry, is often considered beneficial for the later maturation of whisky. This previous use will change the composition of extractives by removing compounds but may also cause the direct or indirect formation of new compounds in the wood, which may then be available for later extraction during whisky maturation. However there is no published, conclusive evidence for claims that the flavour and colour of the whisky is improved by the previous storage of sherry.

Despite the uncertainty over its importance attempts to simulate previous sherry storage have been made. Wine-treatment of casks involves allowing the wood to absorb a very sweet, dark sherry under pressure. Changes in the analytical composition of casks by this treatment include an increase in total esters, colour and sugars, but this appears to be of little significance relative to flavour (Philp, 1989a). Attempts to simulate true sherry shipping cask flavour include processes involving wine, steam and ammonia treatments. The methods include a combined rejuvenation and sherry treatment process, which involves identifying and cleaning European oak casks, followed by adding sherry to the cask interior and allowing it to penetrate the wood (Philp, 1989b).

Other wood treatments

A variety of wood treatments have been experimented with, mostly with the aim of increasing the speed of maturation for various alcoholic beverages. Litchev (1989) described how treating wood with pressurized oxygen at high temperature increased the maturation speed of wine. Maga (1989b) also describes a number of treatments, including ultrasound, which have been claimed to enhance the release of lignin and amino acids, while heating brandy in the barrel combined with ultrasound apparently reduced maturation time from 3 years to 3 months. Both ultraviolet irradiation and the use of γ-radiation have also been reported as enhancing maturation.

Conditions of extraction and maturation

Maturation and extraction results may vary under different conditions, with a wide range of factors influencing the process. For this reason one must be cautious in comparing different studies and in interpreting their relevance to the maturation process. Conditions of maturation can affect both the direct extraction of compounds and subsequent reactions.

Temperature

It is unlikely that any optimum temperature exists in regard to maturation, as the effect on flavour will differ for each of the components (Reazin, 1983). It has been found (Reazin, 1983; Nykänen, 1986) that extraction and formation of flavour congeners will generally occur more rapidly at 30°C compared to 20°C. The greatest increase was in acetaldehyde and fixed acids, while lactones and furfural levels appeared unaffected. Philp (1989a) summarized previous studies that showed how non-volatile components and component groups were significantly influenced by temperature and to a lesser extent by humidity. Maturation of whisky normally occurs at relatively low temperatures: which are used primarily to reduce the rate of evaporation from the cask.

Extraction solvent and proof of distillate

Most studies use either an ethanol-water mixture or raw distillate to extract wood compounds. Nykänen *et al.* (1985) found that maximum extraction efficiency occurred at about 60 per cent alcohol concentration. However, Maga (1989a) found that maximum lactone extraction was obtained with 40 per cent alcohol, while Puech (1984) found that the optimum extraction levels of tannins and lignin by Armagnac occurred at 55 per cent. It is likely that similar variation may be found for other types of extractives. If the alcohol content rises above approximately 60 per cent, then the rate of extraction for colour, solids, tannins, and volatile acids have all been found to decrease. A number of authors have emphasized the effect that distillate strength can have on maturation speeds (for example Baldwin *et al.* 1967; Baldwin and Andreasen, 1974; Sharp, 1983). The reason for this variation is that while the hydrolytic reactions, such as the breakdown of polymeric material, require water, the solubility of degraded compounds improves with increasing alcohol concentration. Therefore the highest rate of extraction will occur at the concentration when these two processes are optimally balanced.

Maga (1989a) found that the pH of the extraction solution affected the degree of lactone extracted from wood by an alcohol solution. Peng *et al.* (1991) described how water extractions were more efficient at higher pH levels. The solution pH will also be influenced by the cask wood extractives, with acidification being characteristic of pyrolysed wood (Sarni *et al.*, 1990). Maga (1989b) described how the pH declines during maturation of wine in wooden barrels, and this may in turn influence the formation of many compounds.

Oxygen content and availability

Oxidative reactions have been intensivily studied in the process of wine fermentation and maturation (see the review by Singleton, 1987). In wine maturation slow oxidation is preferred, and the varying rates at which oxidation will occur in a wooden cask are deemed beneficial as this leads to a more complex mixture of oxidative products than would a constant rate throughout. Sefton (1991), in discussing the maturation of wine, claimed that controlled oxidation led to less astringency (perhaps due to the oxidation of tannins) and increased colour and stability.

Chatonnet (1991) found that old barrels lose their oxidative properties, possibly due to the loss of hydrolysable tannins that may act as catalysts for many oxidative reactions during maturation. Metal ions, such as copper, have also been identified as oxidation catalysts.

The extraction of oak wood using alcohol–water mixtures with varying oxygen concentrations has shown that a higher oxygen content results in greater concentrations of lignin degradation products, such as vanillin and syringaldehyde, while more sinapaldehyde was produced when extracts were prepared with little oxygen (Maarse and van der Berg, 1989). Higher levels of eugenol and furfural were also found in high-oxygen containing extracts. However, despite the lower levels of most lignin degradation products, the extracts obtained with little oxygen had a more harmonic, cognac-like and less astringent flavour.

An indication of the importance of oxygen in whisky maturation is that if a maturing cask is wrapped in a film impervious to air, no change in flavour will take place (Jago personal communication). However, although the exclusion of oxygen is the most likely cause of this result, the treatment will also prevent evaporation and possibly decrease the penetration of the cask by the whisky distillate.

Humidity

This has been claimed to affect the rates of oxidation and particularly evaporation (Castelli and Peynaud, 1990). Guymon and Crowell (1970) described how humidity affects the final proof of maturing brandy. Low humidity produces an increase in alcohol content due to the evaporation rate of water being higher than that of alcohol. In high humidity conditions, the proof declines, as more alcohol than water evaporates (alcohol evaporation being independent of humidity). Evaporation may then result in further effects, due to the changes in the proof of the maturing distillate.

Study methods

Limitations of previous studies

The difficulty in summarizing the effects of oak wood on the maturation of alcoholic beverages reflects the diverse range of disciplines and subjects that it involves, and the numerous approaches and motivations behind the studies undertaken. One of the main aims of this review has been to draw together the various aspects of different research relevant to the subject. An additional problem arises from the various commercial interests involved, from cooperages that do little to discourage various rumours and claims regarding the benefits of cask treatments or wood types, to the producers of the alcoholic products, who while wishing to maintain an aura of mystique around the process of maturation, have powerful financial interests in any understanding that would allow better control over flavour or quicker maturation times. Then there are the many practical limitations and problems arising from the subjective nature of flavour trials and the cost and time involved in research.

It is perhaps not surprising therefore that there is often a notable lack of clarity in many of the studies that have examined the process of whisky maturation. Each study tends to focus on a single factor that may influence the maturation process, while frequently failing to control variation of other factors adequately. Studies on the effect of charring will often compare charred and uncharred casks, but the past use and origin of these casks is frequently uncertain. In regards to the origin of cask wood in particular, there is often little attempt to discover even the botanical species involved, let alone more precise origins. Even less consideration is normally given to the wood age, treatment and seasoning. For example, comparisons between the levels of lactones in different oak types failed to ensure the wood samples were of similar age, despite it being known that the formation of lactones is highly dependent upon the wood age, both of the once living tree and since felling.

Another feature that is frequently ignored, but is always a problem when comparing different studies, is the method of extraction. Many studies involve the extraction of compounds from the wood by either the unmatured distillate (which may vary in its composition), or by using water-ethanol mixtures or other solvents. Both the solvent and the conditions under which the extractions occur influence the resulting composition of the extractive.

Lack of replication is also a frequent problem in many studies, particularly in comparing different sources of oak wood, such as in Puech (1984) and Miller *et al.* (1992). Practical limitations frequently make a high degree of replication impossible for the types of study undertaken.

Generally there has been a lack of planning in many approaches, without adequate ground work having been done to allow practical interpretation of many results in respect to their relevance to the maturation process. Finally, there has been little testing of hypotheses, with work tending to be simply comparative analyses or attempts to correlate various factors. Although possible explanations and hypotheses are generated by these studies, there appears to have been little attempt to follow up with predictive tests.

Priorities for future research

Chemical composition and whisky flavour If any easily applied work is to be undertaken, in comparing different maturation conditions or wood types, it is necessary to know what properties affect the maturation of the whisky. To date, although a wide range of flavour congeners have been identified in whisky, little is known about their relative importance or their specific effects on the flavour of whisky. Most understanding of the importance of congeners is based upon comparing their concentrations with determined flavour thresholds or studying their variation between different whisky types and maturation conditions. However, as has been highlighted by Maga (1985), once cannot rely on individual flavour thresholds to determine whether a compound contributes to the taste, as these thresholds may vary due to interaction between compounds. Furthermore, although comparative analytical studies of different whisky types may allow some indication as to which compounds are primarily involved, these studies would be of more value if the important flavour congeners had already been identified.

As stated by numerous authors (e.g. Paterson and Piggott, 1989; Swan and Howie, 1985; Maarse and van der Berg, 1989) the problem at present is relating trace chemical concentrations with sensory data on flavour and aroma. Ideally, in order to identify important flavour congeners, studies that examine the variation of chemical composition across a specific flavour gradient are required. This would involve the selection of identifiable flavour characteristics, which may be defined as having several levels or varying intensities in different whiskies. Multivariate analysis methods could be used to identify chemical characteristics of the whisky that correlate with the chosen flavour character. As long as a suitably large sample size is used such methods do not require that other flavours be kept constant. The main limitation is the subjective nature of the measurements and whether the chosen flavour is of consequence. However, such difficulties are common to all scientific attempts to study flavour and have been detailed by numerous authors (for example Piggott, 1988; Burgard and Kuznicki, 1990; Lyon *et al.*, 1992).

Derivation of flavour congeners Once flavour congeners of importance have been identified, the next step is to identify those compounds that derive from, or are influenced by the cask wood. Previous studies as well as present knowledge in regard to the chemistry of wood and the maturation process may assist. The eventual aim is to trace the derivation of required flavour congeners back through the series of factors that may influence their appearance in the whisky. First the environmental conditions of the maturation process, and how these influence the levels of congeners, may be studied. The effect of different cask treatments may then be compared before comparison of the wood type and origins is undertaken. This has the benefit of focusing initially on those stages most easily controlled by whisky producers before considering factors less easily determined.

Many of the studies that have produced the most interesting and applicable results have compared the amounts of wood derived compounds found in whisky matured under different conditions or using different types of casks (for example Baldwin *et al.*, 1967; Guymon and Crowell, 1970). Although the relevance of such studies may be easily recognized, they suffer from a number of limitations. The most obvious restrictions on their use is the length of time and the resources needed to carry out such a comparison. It is also often difficult to control the various factors that may influence the properties of the cask. In order for more precise comparisons a swifter method of determining the effect of the wood on the maturation of whisky is required.

Two different developments would be of use in determining the origins of flavour congeners. First there is the need for simple model systems that allow the simulation of whisky maturation under controlled environmental conditions. Such a system would allow the control of temperature and oxygen availability during the storage of a standard raw distillate with the addition of individual or mixtures of oak extractives in known concentrations. This would allow carefully controlled studies on the development of congeners during maturation of whisky under varying conditions. Commercial oak extract is already available and methods exist for purification of many wood compounds thought to influence flavour.

The second need is the development of standard methods of measuring wood properties, to allow better comparisons between different cask treatments or oak types. Recent papers have compared different methods of extracting and measuring compounds (Maga, 1989b; Puech *et al.*, 1990; Scalbert, 1992a) and have shown how they may give widely differing results. It would be useful to develop more standard methods and determine how different methods of measuring wood extractives and flavour relate to the actual performance of a wood type in whisky maturation. Such methods should take account of any physical properties of the wood that might influence extraction or the conditions of maturation.

Conclusions

A suitable supply of cask wood must meet three separate criteria:

1 Economic feasibility—the wood supply must be adequate to meet present and future demand at an economic price.
2 Cooperage criteria—the wood must allow the construction of a tight cask, allowing minimum leakage, and be of suitable strength.
3 Flavour criteria—the wood must have the necessary properties that will produce mature whisky of the desired flavour.

Because of the ability to obtain the required flavour by means of blending and also because flavour properties may to some extent be instilled by cask treatment, there has been a past emphasis upon the other two criteria. However, advances in the understanding of the maturation process and the role of the cask in producing flavour congeners have emphasized the importance of the wood in affecting the flavour of whisky. It is now accepted that the influence of the botanical species of oak on the matured whisky, although modified by cask treatment and age, is undoubtably a major factor. Furthermore the suitability of wood may vary not only between species but also between different geographical regions. The causes of this variation have received little attention, but studies on the variation of wood properties indicate that silvicultural, environmental, and genetic factors may all be important.

One would ideally like to characterize mature whisky flavours, by means of the levels of the various flavour congeners involved. By study of the synthesis or source of these compounds, one could elucidate properties required in the virgin wood, separately from those that may be induced at a later stage through cask treatments or blending of matured distillates. However, although the understanding of the maturation process has improved greatly over recent years, it is still not possible to define a desirable whisky flavour by means of flavour congener levels, let alone construct an index of properties required in the used cask or in the timber from which it is constructed. Furthermore, the role that anatomical features may play in determining the availability of extractive content is not well understood.

The cooperage requirements of wood have also received little attention, although preferences for certain types and characters do exist within the industry. Such requirements will influence the search for new sources of oak wood by imposing additional criteria for suitable wood. It is also apparent that the method of cooperage may greatly influence the flavour derived from the cask. This may allow certain properties of the wood to be altered after harvesting, according to the required needs.

Although the cost of new casks is often considered to be prohibitively high, this must be contrasted with the potential of much more rapid maturation and distinctive flavours that certain types of new casks may offer. However, to utilize this potential requires a better understanding of the way in which wood may influence flavour. Only when swifter, and more predictive methods of estimating flavour effects have been developed will it be possible to examine more fully the variation in the effects of cask and wood types.

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