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KINETICS OF IMPREGNATION/EVAPORATION AND RELEASE OF PHENOLIC COMPOUNDS FROM WOOD TO BRANDY IN EXPERIMENTAL MODEL

CINÉTICAS DE IMPREGNAÇÃO/EVAPORAÇÃO E TRANSFERÊNCIA DE COMPOSTOS FENÓLICOS DA MADEIRA PARA A AGUARDENTE EM MODELO EXPERIMENTAL

Sara Canas[1], A. Pedro Belchior[1], Ana M. Mateus[1,2], M. Isabel Spranger[1], R. Bruno-de-Sousa[3]

[1] Estação Vitivinícola Nacional. INIA. 2565-191 DOIS PORTOS. Portugal.
E-mail:inia.evn.quim@oninet.pt
[2] Aquisição de serviços
[3] Instituto Superior de Agronomia - Departamento de Química Agrícola e Ambiental. Tapada da Ajuda. LISBOA.Portugal
E-mail: bruno@reitoria.utl.pt

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SUMMARY

An essay was carried out under model conditions, simulating the natural ageing of brandies in 250 dm^3 wooden barrels, to study the kinetics of impregnation/evaporation of brandy and the release of phenolic compounds from wood into the brandy. The influence of wood botanical species (Limousin oak and Portuguese chestnut), toasting level (light and strong) and ageing time (0, 42 and 250 days) was investigated. In parallel, a reduced essay with red wine was made to compare the kinetics of impregnation/evaporation of wine and brandy.

The great loss of wine or brandy and the highest extraction of phenolic compounds occur in the first month of the ageing process. Brandy and wine show different kinetics of impregnation/evaporation that seem to be governed by the impregnation rate, in the beginning mostly dependent on water content (15 days) and then mainly determined by their non-volatile composition. The total loss of brandy *per* year is more pronounced than wine (5–15% *versus* 5-9%) and the corresponding impregnated layer is about two-fold thicker.

In spite of the significant effect of wood botanical species, toasting level and ageing time on total phenolic compounds and color intensity of wood and brandy, as verified in previous works, it is also evident the strong influence of wood intraspecific variability (differences between trees and in heartwood age) and toasting procedure variability.

Key words: brandy, woods, ageing, impregnation, physical and chemical characteristics

Palavras-Chave: aguardente, madeiras, envelhecimento, impregnação, características fisico-químicas

INTRODUCTION

The knowledge of the phenomena that occur in the wood during the ageing of beverages has been a reason for many research studies.

Some works (Ribéreau-Gayon *et al.*, 1976; Feuillat *et al.*, 1994) have been made to evaluate the loss of liquid during the ageing process, resulting both from impregnation and volatilization, in wine and in model solutions.

Other important related phenomenon is the release of extractable compounds, especially phenolic compounds, from the wood into the liquid, which has considerable repercussions on its physical-chemical characteristics and sensorial properties (Guymon and Crowell, 1968; Viriot *et al.*, 1993).

Several papers have been focused on the extraction kinetics of phenolic compounds in wines (Rous and Alderson, 1983; Kadim and Mannheim, 1999), in whiskies (Baldwin and Andreasen, 1974; Mosedale, 1995) and in model solutions (Moutounet *et al.*, 1989; Puech *et al.*, 1996; Kadim and Mannheim, 1999), but few studies were made with brandies (Puech *et al.*, 1985).

The extraction of compounds from oak barrels into brandy depends on the pool of potential extractable compounds originally present in the oak, on the composition of the liquid and on the ageing conditions. Thus, this mechanism is conditioned by a large number of interdependent factors, namely the botanical species and the geographical origin of the wood (Guymon and Crowell, 1970; Miller *et al.*, 1992; Marco *et al.*, 1994; Mosedale and Ford, 1996; Mosedale and Savill, 1996; Canas *et al.*, 2000c), the heartwood age (Peng *et al.*, 1991; Klumpers *et al.*, 1994; Viriot *et al.*, 1994), the heat treatment (Nishimura *et al.*, 1983; Artajona *et al.*, 1990; Rabier and Moutounet, 1990; Sarni *et al.*, 1990; Hale *et al.*, 1999; Canas *et al.* 2000b), the relative proportion of water and ethanol (Guymon and Crowell, 1970; Baldwin and Andreasen, 1974; Singleton, 1995), the container size (Guymon e Crowell, 1970; Kadim and Mannheim, 1999) and the warehouse conditions (Cantagrel *et al.*, 1992; Mosedale, 1995; Singleton, 1995).

The objective of this work was to monitoring the loss of brandy during 250 days, under model conditions, simulating the natural ageing of brandies. It was also performed the physical and chemical analysis of woods and corresponding brandies after 42 days and in the final of the period. In parallel, a reduced essay with red wine was made to compare the kinetics of impregnation/ evaporation of wine and brandy.

MATERIAL AND METHODS

Experimental design

This study was based in a factorial experiment, with three factors - wood botanical species, toasting level, with two replications, and ageing time.

The essay was carried out from 18th October 2000 to 26th June 2001.

Materials

Wood

Two heartwood staves of Limousin oak wood (*Quercus robur* L.) - CFL and two heartwood staves of Portuguese chestnut wood (*Castanea sativa* Mill.) - CAST were supplied by a cooperage industry - JMA Gonçalves, where they were seasoned in the open air and toasted. One stave of each wood was submitted to light toasting (QL) and the other was submitted to strong toasting (QF). After that they were cut into pieces with 110 mm long, 110 mm width and 27 mm thickness. Two pieces of each wood x toasting level constitute the control group and the others were used to construct the experimental units, with a glass piece (Figure 1), which had been developed respecting the ratio

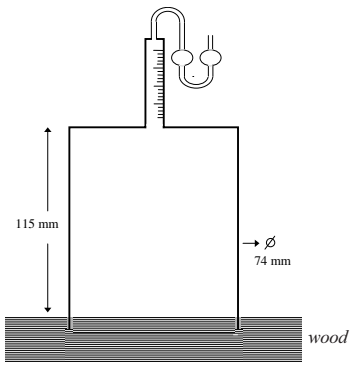


Fig. 1 – Experimental unit scheme (longitudinal plan).
Esquema da unidade experimental (plano longitudinal).

surface area/volume of a 250 dm^3 wooden barrel (86 cm^2/dm^3). The linkage between glass and wood was made with silicone - food quality.

Brandy

32 experimental units were filled with the same Lourinhã brandy (485 cm^3) and placed at the cellar of Estação Vitivinícola Nacional, in similar environmental conditions, to simulate the enological practice.

The experimental units of wood x toasting level were identified as CAST QF, CAST QL, CFL QF and CFL QL.

Wine

Three experimental units (CAST QF, CFL QF and CFL QL) were filled with the same red wine, produced at Estação Vitivinícola Nacional, and were placed near the brandy's experimental units.

Methods

Measurement of the liquid volume

The measurements of the brandy and wine volumes into the experimental units were performed in mm and converted into cm^3. The frequency of measurements was: daily during the first ten weeks, twice a week for 16 weeks and then weekly for 20 weeks (250 days). Whenever necessary the experimental unit was refilled with brandy or wine to the original volume.

The kinetic curves were drawn from the mean values of liquid loss of three to five experimental units.

Cellar temperature and relative humidity were also monitoring during the essay.

Wood and brandy sampling

T (0 days) - eight wood pieces (control group) were cut into layers of two mm from the inner surface to outer surface and the two-mm-thick pieces of wood were then grounded in a hammer-mill (Wiley). The corresponding wood powder was immediately analyzed. The brandy (control) was also analyzed.

T1 (42 days) and T2 (250 days) – two experimental units of each wood x toasting level were randomly selected. The unit was emptied and the brandy was homogenized and sampled. Then the unit was destroyed and the wet layer of the wood piece was measured and cut. The wood chips were grounded, homogenized, weighed and the sample was kept.

Wood extraction

Maceration of wood powder (1g) was carried out under rotary agitation for 180 min at 20 ºC, with 20 cm^3 of an ethanol/water solution (55:45 v/v and pH 4,2 adjusted with hydrochloric acid) as described by Caldeira *et al.* (1996). The wood extracts were filtered through a glass microfibre filter (Whatman GF/C) on a Büchner funnel.

Analytical methods

Color intensity (A440) – measurement of absorbance at 440 nm (Belchior e Carvalho, 1983).

Total polyphenol index (Ipt) – product of the measured absorbance at 280 nm by the dilution factor (Ribéreau-Gayon, 1970).

All measurements were carried out in a Shimatsu UV-265 spectrophotometer using a quartz cell of 0,1 cm path length, and were referred to 1 cm path length.

Dry extract weight – usual method of OIV (1994).

Statistical analysis

The two-way analysis of variance and the correlations analysis were performed using Statgraphic v.5 (STSC inc., Rockville, USA).

RESULTS AND DISCUSSION

Kinetics of Impregnation/evaporation

The kinetics of impregnation/evaporation of brandies shown in Figure 2 (b) represent a three-stage process: during the initial phase (15 days) there is a slow increasing of the brandies loss, followed by a sharp increasing (30 days)

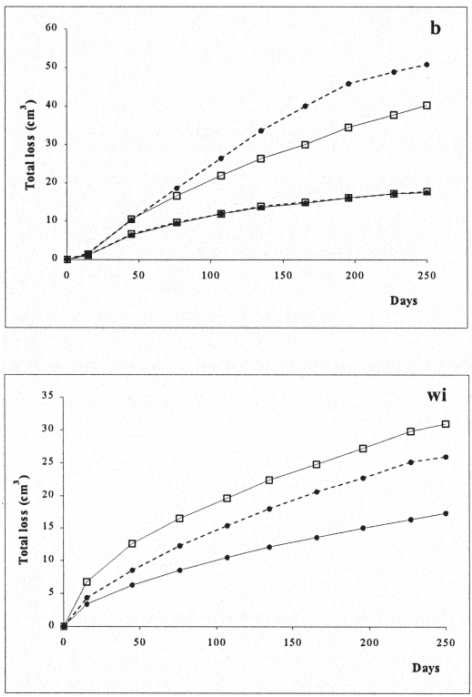


Fig. 2 - Kinetics of impregnation/evaporation according to wood x toasting level in: (b) brandies and (wi) wines.
CAST QF; CAST QL; CFL QF; CFL QL.
Cinéticas de impregnação/evaporação de acordo com madeira x nível de queima em: (b) aguardentes e (wi) vinhos.

and, finally, during the third stage the increment trends to diminishing. The brandies ageing in CFL QL has a slightly different behavior, with a more pronounced second phase.

In wines (Figure 2 wi) the kinetics curves seem to imply only a two-stage process, starting with a strong slope and then trends to diminishing.

The main difference between brandies and wines is their behavior in the initial stage that could be explained by their water content (88% in wine *versus* 23% in brandy). The faster penetration through the wood of water than ethanol (Guymon e Crowell, 1970; Singleton, 1995) causes a more rapid loss of wine than brandy.

Since the water/ethanol content of wine and brandy is practically constant during the essay, the total loss of wine is much lesser than of brandy – 5 to 9% versus 5 to 15%, the wet layer found in woods that had contacted with wine (3–4,5 mm) is thinner than in woods that had contacted with brandy (8–9 mm), then the process seem to be governed by the impregnation rate, which is mostly dependent on the water content in the beginning (15 days) and then is mainly limited by the non-volatile composition (Mosedale, 1995) and probably facilitated by the alcohol content of wine or brandy.

Although Feuillat *et al.* (1994) had reported the influence of warehouse temperature and humidity on the total loss of liquid, the kinetics of impregnation/evaporation obtained do not reveal this fact. This discrepancy can be justified by: the lower thermal amplitude of our cellar (Figure 3); the type of the liquid

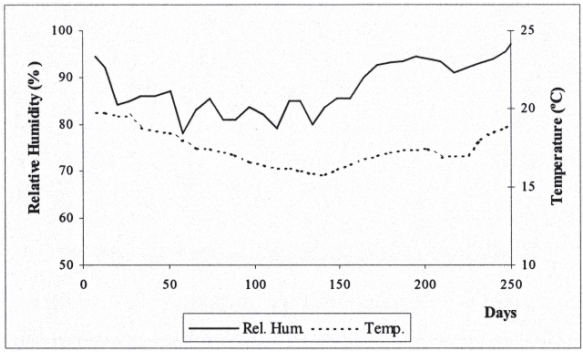


Fig. 3 – Warehouse temperature and relative humidity during the essay.
Temperatura e humidade relativa da cave durante o ensaio.

used to fill the experimental units; the thickness of the wood pieces (9 mm *versus* 27 mm); the intraspecific variability of wood.

The results obtained (Figure 2 b) in brandies show the strong influence of intraspecific variability of wood that overlaps the effect of the other factors,

such as botanical species (Feuillat *et al.*, 1994) and toasting level. So, it is not evident the influence of the toasting intensity on the wood porosity and accessibility of brandy to wood extraction sites, as reported by Sarni *et al.* (1990). Also the differences of permeability between oak and chestnut woods, derived from their specific anatomical features – types of ray parenchyma and abundance of tyloses (Feuillat and Keller, 1997; Vivas *et al.*, 2000), are not clear.

Extraction of phenolic compounds

The results of variance analysis (Table I) show a very significant effect of the wood botanical species on the content of phenolic compounds of wood and brandy, as well as on the dry extract of brandies. Chestnut wood is the richest

TABLE I

Effect of the botanical specie, toasting level and ageing time on the physical and chemical characteristics of woods and brandies

Efeito da espécie botânica, nível de queima e tempo de envelhecimento nas características físico-químicas das madeiras e das aguardentes

		Wood		Brandy		
		Ipt	**A440**	**Ipt**	**A440**	**Dry extract**
Botanical specie						
effect		**	*	**	**	**
CAST	*x*	178,97***b***	1,02***a***	15,69***b***	0,18***b***	1,15***b***
	SD	40,532	0,476	13,458	0,179	0,909
CFL	*x*	75,58***a***	1,17***b***	5,70***a***	0,11***a***	0,91***a***
	SD	21,680	0,496	5,922	0,143	0,698
Toasting level						
effect		**	**	*n.s*	**	*n.s*
QF	*x*	115,10***a***	1,40***b***	11,76	0,23***b***	0,99
	SD	38,781	0,517	11,021	0,184	0,780
QL	*x*	139,45***b***	0,79***a***	9,62	0,06***a***	1,07
	SD	78,211	0,124	12,108	0,070	0,856
Ageing time						
effect		*n.s*	**	**	**	**
T	*x*	126,58	1,21***b***	0,00***a***	0,00***a***	0,00***a***
	SD	72,222	0,482	0,000	0,000	0,000
T1	*x*	136,29	1,27***b***	11,99***b***	0,19***b***	1,30***b***
	SD	58,097	0,611	7,373	0,161	0,247
T2	*x*	118,96	0,80***a***	20,09***c***	0,24***b***	1,80***c***
	SD	60,992	0,088	11,714	0,153	0,293

Botanical specie and toasting level: x = mean of 12 values; Ageing time: x = mean of 8 values. Means followed by the same letter in a column are not significantly different at 0,05 level of significance; SD = standard deviation; ns = without significant difference.*

A440 - color intensity; Ipt - total polyphenol index.

in those compounds, as already verified (Canas *et al.*, 2000c). The corresponding brandies are also richer in phenolic compounds and dry extract than those aged in oak wood.

The color intensity of the brandy is very significantly affected by the wood botanical species, while the color intensity of the wood is only significantly affect by this factor. The weak correlation between color intensity of wood and brandy is probably explained by the different behavior of CFL QF wood (Figure 4), which causes some distortion of the mean value of wood color intensity.

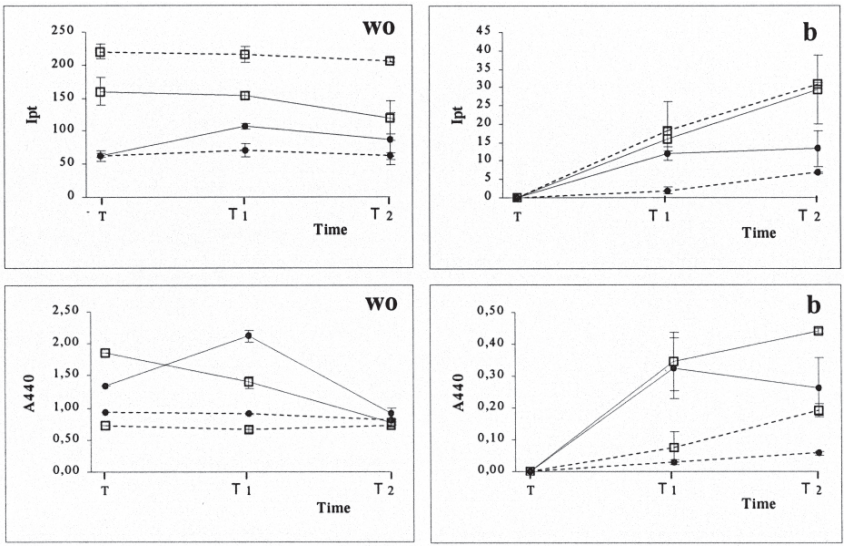


Fig. 4 – Evolution of total phenolic content and color intensity of: (wo) woods and (b) brandies.
CAST QF; CAST QL; CFL QF; CFL QL.
Evolução dos polifenóis totais e da intensidade da cor de: (wo) madeiras e (b) aguardentes.

Higher color intensity of brandies aged in chestnut than in oak wood is coherent with that observed in three-year-aged brandies in 250 dm^3 wooden barrels (Canas *et al.*, 2000a). It can mean that this trend becomes evident immediately from the first month of ageing, revealing the great importance of extraction and oxidation processes in this early period.

Toasting level has a very significant influence on the total content of phenolic compounds in wood and on the color intensity of wood and brandy (Table I). The woods with light toasting show the highest content of phenolic compounds, but the lowest color intensity, while the corresponding brandies present lower content of phenolic compounds and lower color intensity than the brandies aged in wood with strong toasting. Brandies cannot be distinguishing by their dry extract as a function of wood toasting level, but light toasting induces higher dry extract.

The results of total phenolic content in woods do not follow the general tendency that QF>QL (Canas *et al.*, 2000b).

According to the toasting level, differentiation of brandies by their color intensity is in agreement with that observed in a previous work (Canas *et al.*, 2000a).

It is reliable that the content of phenolic compounds in wood is affected by the behavior of CFL QF wood. On the other hand, the results obtained could be partially explained by the variability of wood toasting procedure (Chatonnet and Boidron, 1989; Artajona *et al.*, 1990; Chatonnet *et al.*, 1993; Chatonnet, 1999; Canas *et al.*, 2000b) and by the intraspecific variability of wood anatomical characteristics (Polge and Keller, 1973; Deret-Varcin, 1983; Mosedale *et al.*, 1996b) and chemical composition (Peng *et al.*, 1994; Viriot *et al.*, 1994; Masson *et al.*, 1995; Mosedale *et al.*, 1996a,b; Feuillat *et al.*, 1997), owing to differences between trees and in heartwood age.

Except the total phenolic content of wood, the studied parameters of woods and brandies depend largely on the ageing time (Table I). It is observed that increasing ageing time increases these parameters in brandy and decreases wood color intensity.

The ageing time effect is also conditioned by the behavior of CFL QF wood, which causes the increase of wood total phenolic content and color intensity at T1, while the tendency is to diminishing with time. Similarly, the evolution of CFL QF brandy affects the mean value of brandies color intensity.

It is interesting to point out the synchronism of the decreasing trend of total phenolic content in woods and its increasing in brandies with the ageing time (Figure 4), as a consequence of the release of phenolic compounds from the wood to the brandy. This fact is also supported by the significant correlation between them.

The more pronounced extraction of phenolic compounds occurring in the first month (T1) of the ageing process could derive both from the higher availability of those compounds in wood surface and the higher gradient between wood and brandy, which facilitate their transfer into the brandy (Kadim and Mannheim, 1999). On the other hand, the higher impregnation/evaporation rate verified during this period could also contribute to that phenomenon.

During the second stage (T2) the total phenolic content of brandy is probably determined by the resistance to mass transfer on the wood side.

In a methodological point of view the experimental model satisfying the proposed objectives. However, a rigorous control of the wood must be paid in order to eliminate the highest number of variability factors. It means that wood staves that will be used in future essays must be obtained from the same tree and from the same position in the trunk.

CONCLUSIONS

The great loss of wine or brandy and the uppermost extraction of phenolic compounds occur in the first month of the ageing process.

Brandy and wine show different kinetics of impregnation/evaporation that appear to be governed by the impregnation rate, which is initially dependent on their water content (15 days) and then mainly determined on their non-volatile composition.

There is a higher loss of brandy *per year* (5–15%) than wine (5-9%), and the corresponding wet layer in woods that had contacted with brandy (8–9 mm) is thicker than in those that had contacted with wine (3–4,5 mm).

Although the significant effect of wood botanical species, toasting level and ageing time on the content of phenolic compounds and color intensity of wood and brandy, it is evident the strong influence of wood intraspecific variability and toasting procedure variability.

The results are in agreement with those obtained in enological practice.

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RESUMO

Cinéticas de impregnação/evaporação e transferência de compostos fenólicos da madeira para a aguardente em modelo experimental

Com o objectivo de estudar as cinéticas de impregnação/evaporação da aguardente e a transferência de compostos fenólicos da madeira foi realizado um ensaio em condições modelo, simulando o envelhecimento natural de aguardente em vasilhas de madeira de 250 dm^3. Foi analisada a influência da espécie botânica da madeira (carvalho Limousin e castanho português), do nível de queima (ligeiro ou forte) e do tempo de envelhecimento (0, 42 e 250 dias). Através de um ensaio paralelo, em pequena escala, utilizando vinho tinto, procurou-se comparar as cinéticas do vinho e da aguardente.

A grande perda de aguardente e de vinho e a maior extracção de compostos fenólicos da madeira pela aguardente ocorrem logo no primeiro mês de envelhecimento. A aguardente e o vinho apresentam cinéticas de impregnação/evaporação diferentes, que parecem ser determinadas pela taxa de impregnação, inicialmente dependente sobretudo do seu teor de água (15 dias) e depois principalmente condicionada pela sua composição não-volátil. A perda total anual de aguardente é mais acentuada que a de vinho (5–15% *versus* 5-9%) e a correspondente camada de madeira impregnada é cerca de duas vezes mais espessa.

Apesar do efeito significativo da espécie botânica, do nível de queima e do tempo de envelhecimento no teor de compostos fenólicos e na intensidade da cor de madeiras e aguardentes, como verificado em trabalhos anteriores, torna-se bastante evidente a influência da variabilidade intraespecífica da madeira (associada às diferenças entre árvores e à idade da madeira) e da variabilidade da queima.

RÉSUMÉ

Cinétiques d'imprégnation/évaporation et transfert des composés phénoliques du bois pour l'eau-de-vie en modèle expérimental

Ce travail consiste en l'étude des cinétiques d'imprégnation/évaporation de l'eau-de-vie et le transfert de composés phénoliques du bois, en conditions modèle, avec un dispositif expérimental, simulant le vieillissement naturel de l'eau-de-vie en fût de bois de 250 dm^3. L'influence de trois facteurs est analysée: l'espèce botanique (chêne Limousin et châtaignier portugais), le niveau de brûlage (léger et fort) et le temps de vieillissement (0, 42 et 250 jours). Un essai parallèle, moins complet, a été conduit sur du vin rouge pour comparer les cinétiques de ce vin et celles de l'eau-de-vie.

La plus grande perte en eau-de-vie et en vin et la plus grande extraction des composés phénoliques du bois par l'eau-de-vie ont lieu très rapidement pendant le premier mois de vieillissement. L'eau-de-vie et le vin présentent des cinétiques d'imprégnation/évaporation différentes qui semblent être déterminées par le taux d'imprégnation. L'imprégnation initiale dépend surtout de la teneur en eau (15 jours) et après de la fraction non volatile. La perte totale annuelle de l'eau-de-vie est plus importante que celle du vin (5-15% contre 5-9%), ce qui correspond a une couche de bois imprégnée environ deux fois plus importante.

Bien que l'effet de l'espèce botanique, du niveau de brûlage et du temps de vieillissement sur les composés phénoliques totaux et sur l'intensité de la couleur soit significatif, comme déjà vérifié dans des travaux précédents, les résultats sont également fortement influencés par la variabilité intra spécifique du bois (inter arbres et âge du bois de cœur) et par la variabilité du brûlage.

REFERENCES

Artajona J., Barbero E., Llobet M., Marco J., Parente F., 1990. Influence du "bousinage" de la barrique sur les qualites organoleptiques des brandies vieillies en fûts de chêne. *In: Les eaux-de-vie traditionnelles d'origine viticole*. 197-205. Bertrand, A. (ed.), Lavoisier - Tec & Doc, Paris.

Belchior A.P., Carvalho E., 1983. A cor em aguardentes vínicas envelhecidas: método espectrofotométrico de determinação e relação com os teores em fenólicas totais. *Ciência. Tec. Vitiv.*, **2**, 29-37.

Caldeira I., Canas S., Belchior A.P., 1996. Optimização das condições de extracção de aparas de madeira. Relatório interno, Secção de aguardentes, EVN - INIA.

Canas S., Belchior A. P., Caldeira I., Spranger M.I., Bruno de Sousa R., 2000a. La couleur et son évolution dans les eaux-de-vie *Lourinhã* pendant les trois premières années du vieillissement. *Ciência. Tec. Vitiv.*, **15**, 1-14.

Canas S., Grazina N., Spranger M.I., Belchior A.P., 2000b. Modelisation of heat treatment of

Portuguese oak wood (*Quercus pyrenaica* L.). Analysis of the behaviour of low molecular weight phenolic compounds. *Ciência. Tec. Vitiv.*, **15**, 75-94.

Canas S., Leandro M.C., Spranger M.I., Belchior A.P., 2000c. Influence of botanical species and geographical origin on the content of low molecular weight phenolic compounds of woods used in Portuguese cooperage. *Holzforschung*, **54**, 255-261.

Cantagrel R., Mazerolles G., Vidal J.P., Galy B., Boulesteix J.M., Lablanquie O., Gaschet J., 1992. Evolution analytique et organoleptique des eaux-de-vie de Cognac au cours du vieillissement. 2ª partie: Incidence de la température et de l'hygrométrie des lieux de stockage. *In: Elaboration et connaissance des spiritueux*. 573 - 576. Cantagrel R. (ed.), Lavoisier - Tec & Doc, Paris.

Chatonnet P., 1999. Discrimination and control of toasting intensity and quality of oak wood barrels. *Am. J. Enol. Vitic.,* **50**, 479-494.

Chatonnet P., Boidron J.N., 1989. Incidence du traitement thermique du bois de chêne sur sa composition chimique. 1ere Partie: Définition des paramètres thermiques de la chauffe des fûts en tonnellerie. *J. Intern. Sci. Vigne Vin*, **23**, 77-87.

Chatonnet P., Boidron J.N., Dubourdieu D., 1993. Maîtrise de la chauffe de brûlage en tonnellerie. Application à la vinification et à l'élevage des vins en barriques. *Rev. Fr. Oenol.*, **144**, 41-53.

Deret-Varcin E., 1983. Etude de la qualité du bois de trois types de chênes (rouvres, pédunculés et intermédiaires), en forêt de Morimond. *Ann. Sci. For.*, **40**, 373-398.

Feuillat F., Moio L., Guichard E., Marinov M., Fournier N., Puech J.-L., 1997. Variation in the concentration of ellagitannins and cis- and trans-β-methyl-γ-octalactone extracted from oak wood (*Quercus robur* L., *Quercus petraea* Liebl.) under model wine cask conditions. *Am. J. Enol. Vit.*, **48**(4), 509-515.

Feuillat F., Keller R. (1997). Variability of oak wood (*Quercus robur* L., *Quercus petraea* Liebl.). Anatomy relating to cask properties. *Am. J. Enol. Vit.*, **48**(4), 502-508.

Feuillat F., Perrin J.R., Keller R., 1994. Simulation expérimentale de "l'interface tonneau": mesure des cinétiques d'imprégnation du liquide dans le bois et d'évaporation de surface. *J. Int. Sci. Vigne Vin*, **28**, 227-245.

Guymon J.F., Crowell E.A., 1968. Separations of vanillin, syringaldehyde and other aromatic compounds in the extracts of French and American oak woods by brandy and aqueous alcohol solutions. *Qual. Plant. Materiae*, **16**, 302-333.

Guymon J.F., Crowell E.A., 1970. Brandy aging. Some comparisons of American and French oak cooperage. *Wines & vines*, 23-25.

Hale M.D., McCafferty K., Larmie E., Newton J., Swan J.S., 1999. The influence of oak seasoning and toasting parameters on the composition and quality of wine. *Am. J. Enol. Vitic.*, **50**, 495-502.

Kadim D., Mannheim C.H., 1999. Kinetics of phenolic extraction during aging of model wine solution and white wine in oak barrels. *Am. J. Enol. Vit.*, **50**, 33-39.

Klumpers J., Scalbert A., Janin G., 1994. Ellagitannins in European oak wood: polymerization during wood ageing. *Phytochem.*, **36**, 1249-1252.

Marco J., Artajona J., Larremi M.S., Rius F.X., 1994. Relationship between geographical origin and chemical composition of wood of oak barrels. *Am. J. Enol. Vitic.*, **45**, 192-200.

Masson G., Moutounet M., Puech J.-L., 1995. Ellagitannin content of oak wood as a function of species and of sampling position in the tree. *Am. J. Enol. Vit.*, **46**, 262-268.

Miller D.P., Howell G.S., Michaelis C.S., Dickmann D.I., 1992. The content of phenolic acid and aldehyde flavor components of white oak as affected by site and species. *Am. J. Enol. Vitic.*, **43**, 333-338.

Mosedale J.R., 1995. Effects of oak wood on the maturation of alcoholic beverages with particular reference to whisky. *Forestry*, **68**, 203-230.

Mosedale G., Charrier B., Crouch B., Janin G., Savill P.S., 1996a. Variation in the composition and content of ellagitannins in the heartwood of European oaks (*Quercus robur* and *Quercus petraea*). A comparison of two French forests and variation with heartwood age. *Ann. Sci. For.*, **53**, 1005-1018.

Mosedale J.R., Charrier B., Janin G., 1996b. Genetic control of wood colour, density and heartwood ellagitannin concentration in European oak (*Quercus petraea* and *Q. robur*). *Forestry*, **69**, 111-124.

Mosedale J.R., Ford A., 1996. Variation of the flavour and extractives of European oak wood from two French forests. *J. Sci. Food Agric.*, **70**, 273-287.

Mosedale J.R., Savill P.S., 1996. Variation of heartwood phenolics and oak lactones between the species and phenological types of *Quercus petraea* and *Q .robur*. *Forestry*, **69**, 47-55.

Moutounet M., Rabier P., Puech J.-L., Varette E., Barillère J.-M., 1989. Analyse CLHP d'extractibles du bois de chêne. Application à un vin de Chardonnay. *Sci. Aliments*, **9**, 35-51.

Nishimura K., Ohnishi M., Masahiro M., Kunimasa K., Ryuichi M., 1983. Reactions of wood components during maturation. *In: Flavour of distilled beverages: origin and development.* 241 - 255. Piggott, J.R. (ed.), Ellis Horwood Limited, Chichester.

OIV, 1994. *Recueil des méthodes internationales d'analyse des boissons spiritueuses, des alcools et de la fraction arômatique des boissons*, 311 p., OIV, Paris.

Peng S., Scalbert A., Monties B., 1991. Insoluble ellagitannins in *Castanea sativa* and *Quercus petraea* woods. *Phytochem.*, **30**, 775-778.

Polge H., Keller R., 1973. Qualité du bois et largeur d'accroissements en forêt de Tronçais. *Ann. Sci. For.*, **30**, 91-125.

Puech J.-L., Feuillat F., Mosedale J.R., Puech C., 1996. Extraction of ellagitannins from oak wood of model casks. *Vitis,* **35**, 211-214.

Rabier P., Moutounet M., 1990. Evolution d'extractibles de bois de chêne dans une eau-de-vie de vin. Incidence du thermotraitement des barriques. *In: Les eaux-de-vie traditionnelles d'origine viticole*. 220-230. Bertrand, A. (ed.), Lavoisier - TEC & DOC, Paris.

Ribéreau-Gayon P., 1970. Le dosage des composés phénoliques totaux dans les vins rouges. *Chim. Anal.*, **52**, 627-631.

Ribéreau-Gayon J., Peynaud E., Ribéreau-Gayon P., Sudraud P., 1976. *Traité d'oenologie. Siences et techniques du vin*. Tome 3, 719 p., Dunod, Paris.

Rous C., Alderson B., 1983. Phenolic extraction curves for white wine aged in French and American oak barrels. *Am. J. Enol. Vit.*, **34**, 211-215.

Sarni F., Moutounet M., Puech J.-L., Rabier Ph., 1990. Effect of heat treatment of oak wood extractable compounds. *Holzforschung*, **44**, 461-466.

Singleton V., 1995. Maturation of wines and spirits: comparisons, facts and hypotheses. *Am. J. Enol. Vit.*, **46**, 98-115.

Viriot C., Scalbert A., Hervé-du-Penhoat C.L.M., Moutounet M., 1994. Ellagitannins in woods of sessile oak and sweet chestnut. Dimerization and hydrolysis during wood ageing. *Phytochem.*, **36**,1253-1260.

Viriot C., Scalbert A., Lapierre C., Moutounet M., 1993. Ellagitannins and lignins in aging of spirits in oak barrels. *J. Agric. Food Chem.*, **41**, 1872-1879.

Vivas N. (2000). Apports récents à la connaissance du chêne de tonnellerie et à l'élevage des vins rouges en barrique. *Bull. OIV*, **827-828**, 79-108.