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Research Article

Evaluating the vapour evaporation from the surface of liquid pure organic solvents and their mixtures

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Abstract

Some perfumery and cosmetic products represent mixtures and they include large parts of solvents as ethanol, water, acetone and isopropyl alcohol. Solvents as pure hexane and ethanol-water mixtures are used in the solvent extraction of oil-bearing plant materials and herbs. The goal of this study was the emissions of volatile solvents released during above pointed productions to be evaluated. It was found that the specific evaporation rate varies from 1.2 kg/(m².h) (for pure methoxy-propanol) to 66 kg/(m².h) (for three-component mixture including acetone). The evaporation rate is higher for solvents with higher vapour pressure and at a higher velocity of surrounding air. The evaporation is less intensive from pure solvents than their mixtures. The time for the evaporation from a film of solvents and their mixtures was also evaluated. It varies from 14 s (for acetone) to 9 min (for methoxy-propanol). Practical applications: The evaluation of volatile solvent emissions is a mandatory step in the design of plants for manufacturing perfumery, cosmetics, deriving essential and edible oils by means of organic solvents. Most of volatile organic solvents used are highly flammable and healthy hazardous. For assuring fire safety and safety-health working conditions and environment the ventilation system must be designed on the base of these emissions evaluated. The obtained results were applied for plant design of the company “Star Nails Bulgaria” Ltd. Plovdiv for manufacturing cosmetic products.

Keywords: volatile solvent, acetone, isopropyl alcohol, ethanol, hexane

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Introduction

Some perfumery and cosmetic products represent mixtures and they include large parts of solvents as ethanol, water, acetone, isopropyl alcohol and others. Solvents as pure hexane and ethanol-water mixtures are used for extracting oil-bearing plant materials and herbs. These solvents can be referred to liquids with flash point less or equal to 28°C. For instance, the flash point of acetone is -20°C, it is 13°C for isopropyl alcohol, 16.6°C for pure ethanol and 26°C for 40% ethanol-water mixture (Haynes 2014). According to Art. 8 of Bulgarian government regulation I3-1971 (2009) for fire safety, the work areas are classified in the heaviest category F5A for functional fire safety if there are operations with above pointed solvents. For this category, the minimal multiplicities of ventilation in case of normal and emergency modes should be 8 h⁻¹ and 16 h⁻¹, respectively. The organic solvents – what are the objects in this study – represent volatile organic compounds according to Bulgarian government regulation № 7 (2003) and the Council directive 1999/13/EC (1999) of the European Council. Volatile organic compound according to the first regulation is each organic compound whose vapour pressure (at a temperature of 20°C) is more than 10 Pa. For instance, the vapour pressure (at 20°C) of acetone, ethyl acetate, ethanol and isopropyl alcohol is respectively 24.6 kPa; 9.7 kPa; 5.8 kPa; 4.2 kPa (Haynes 2014). The workshops operating with above pointed organic solvents and their mixtures must have proper mechanical ventilation and fire systems for ensuring safe working environment. For the design of these systems the emissions of evaporating solvents should be evaluated quantitatively. The known publications as Hofman (1932) and Mackay and van Wesenbeeck (2014) reported for the evaporation rate of solvents, but only in case of still surrounding air. There are many studies (Carrier 1918; Rohwer 1931; Smith et al. 1994; Poósa and Varju 2017) for water evaporation from free surfaces. They reported many semi-empirical correlations that take into account the velocity of surrounding air.

The goal of this study was to be evaluated the emission of volatile solvents evaporated from the free surface of pure liquid solvents and some their two- and three-component mixtures in case of using mechanical ventilation. The particular tasks were the

evaluation of evaporation rates and the time for evaporating solvents from the free surface of a liquid film including these solvents.

Materials and Methods

Materials

The objects of interest were the following pure organic solvents:

- (i) Ethanol, acetone, isopropyl alcohol, ethyl acetate, methoxy-propanol used as ingredients in the manufacturing of perfumery products and cleaners for nail varnishes;
- (ii) Hexane and ethanol used as mediator agents for extracting edible and essential oils from raw plant materials.

Some selected physical properties of these solvents, collected from Haynes (2014), Poling et al. (2008) and DDBST date bank, are presented in Table 1.

Table 1. Selected physical properties of the solvents studied

Solvents	Molar mass, g/mol	Vapour pressure at 20 °C, kPa	Relative density of vapours, 1	Liquid density, kg/m ³	Boiling point at 101.3 kPa, °C
Methoxy-propanol	90.1	1.45	3.12	998	118.5
Water	18.0	2.31	0.62	922	99.7
Isopropyl alcohol	60.1	4.24	2.08	661	82.6
Ethanol	46.1	5.83	1.60	902	78.24
Ethyl acetate	88.1	9.70	3.05	789	77
n-Hexane	86.2	17.6	2.98	786	68.7
Acetone	58.1	24.6	2.01	784	56.05

Objects of the study were also the following model mixtures of solutions:

- (i) Two-component mixture: ethanol - water that models “Antimicrobial hand gel”. The shares of ethanol and water in this model mixture are respectively 75% and 25%.
- (ii) Two-component mixture: isopropyl alcohol – water that models “Nail gel remover liquid”. The shares of its components are 83% and 17%, respectively.

(iii) Two-component mixture: acetone – water that models “Liquid nail polish remover”. The shares of its components are 95% and 5%, respectively.

(iv) Three-component mixture: acetone – water - ethyl acetate that also models “Liquid nail polish remover”. The shares of its components are respectively 94%, 4% and 2%.

Methods

For evaluating the evaporation rate (kg/h) of solvent from a liquid surface the following well known equation for the two-film model of Lewis and Whitman was used:

$$E = k.F.(x_S - x_A), \quad (1)$$

where F is the area between liquid surfaces and air, m²;

x_S, x_A – the contents of solvent in dry air, kg/kg air;

k - the coefficient of evaporation or mass transfer coefficient, kg/(m².h). For determining this coefficient depending on the velocity of airflow the following well-recognised correlation of Carrier (1918) was employed:

$$k = 25 + 19.v, \quad (2)$$

where v is the velocity of air above the free surface of liquid, m/s.

The correlation of Carrier (1918) for the rate of water evaporation is recommended by the ASHRAE Handbook (Parsons 1999) and by the popular engineering website Engineering toolbox (2018) as well. Physical properties of vapours that influence to mass transfer coefficient k are their mass diffusivity in air, viscosity and density. These properties of the solvents studies and on the other hand the water properties are comparable. For that Eqn. 2 could be also applied for the solvents studied with acceptable accuracy for engineering evaluations.

In case of evaporation from pure solvents the maximal content of solvent x_S in air saturated with solvent vapours and the content of solvent x_A in the surrounding air were determined as follows:

$$x_S = (M_S/M_A).p_S/(p_A - p_S), \quad x_A = 0, \quad (3)$$

where M_S and M_A= 28,96 Da are the molecular masses of solvent and air, respectively, Da; p_S – the vapour pressure of solvent at ambient temperature 20°C, Pa; p_A = 101.3 kPa – the atmospheric pressure of air.

For two-component mixtures of solvents (as ethanol or isopropyl alcohol) with water the solvent content x_S in air saturated with solvent vapours and the solvent content x_A in the surrounding air were calculated as follows:

$$x_S = (M_S/M_A).p_S/(p_A - p_S - p_W), \quad x_A = 0, \quad (4)$$

where p_W is the vapour pressure of water at ambient temperature 20°C, Pa.

For two-component mixtures the maximal water content x_S in air saturated with solvent and water vapours was calculated in a similar way:

$$x_S = (M_W/M_A).p_W/(p_A - p_S - p_W), \quad (5)$$

where M_W = 18 Da is the molecular mass of water.

For these mixtures the water content x_A in the surrounding air was determined as:

$$x_A \approx \varphi.x_S, \quad (6)$$

where φ = 0.7 is the assumed relative humidity of surrounding air.

For three-component mixture, the maximal contents x_{S1} and x_{S2} of the first solvent S1 (acetone) and the second solvent S2 (ethyl acetate) and the content x_A of these solvents in the surrounding air were calculated as follows:

$$\left. \begin{aligned} x_{S1} &= (M_{S1}/M_A).p_{S1}/(p_A - p_{S1} - p_{S2} - p_W), \\ x_{S2} &= (M_{S2}/M_A).p_{S2}/(p_A - p_{S1} - p_{S2} - p_W), \\ x_A &= 0 \end{aligned} \right| \quad (7)$$

where p_{S1}, p_{S2} are respectively the vapour pressures of solvents S1 and S2 at ambient temperature 20°C, Pa;

M_{S1}, M_{S2} - the molecular masses of solvents S1 and S2, respectively, Da.

The maximal water content x_{SW} and the water content x_A in the surrounding air for these mixtures were determined in a similar way:

$$\left. \begin{aligned} x_{SW} &= (M_W/M_A) \cdot p_W / (p_A - p_{S1} - p_{S2} - p_W), \\ x_A &\approx \varphi \cdot x_S. \end{aligned} \right| (8)$$

where $\varphi = 0.7$ is the assumed relative humidity of surrounding air.

For evaluating the solvent evaporation in closed vessels during their prolonged storage the following assumption were done. The velocity in airspace over the liquid solvent in these vessels was assumed as zero. The content of all solvents in the head space of vessel was determined as $x_A \approx \varphi \cdot x_W$, where $\varphi = 0.7$ is the assumed relative saturation of air with solvent vapours.

The evaporation rate was calculated for five levels of the velocity of surrounding air in a range from 0.1 to 1.5 m/s. According to ISO 7730 (2005) for thermal comfort of indoor environment, the velocity of surrounding air should be less than 0.2 m/s in winter and less than 0.25 m/s in summer. ASHRAE Standard 55 (2004) points that the maximum allowable velocity of air in case of emergency ventilation should be less than 1.5 m/s. The studied range from 0.1 to 1.5 m/s is in accordance with Orosa and Oliveira (2012) as well.

Results and Discussion

Specific evaporation rates

These rates for six pure organic solvents from a free liquid surface with an area of 1 m² are presented in Table 2. These results are compatible with the values pointed in previous research (Hofman 1932). The evaporation rate is higher for compounds with higher vapour pressure as the rate for acetone is about 170 times higher than the rate for water evaporation. The evaporation rate in case of emergency mode of ventilation (at a velocity $v = 1.5$ m/s) is about two times higher than the rate in case of normal operation mode (at $v = 0.2$ m/s).

The specific evaporation rates for the free surface of two-component mixtures “ethanol – water”, “isopropyl alcohol – water” and “acetone – water” are presented in Table 2 as well. The evaporation rates of solvents and water were determined separately. The evaporation rates of solvents in these mixtures

are higher than the rate of corresponding pure solvents. These results are in accordance with the observations of other researches (Hofman 1932). The mixture of vapours contains mainly solvent vapours (above 95%).

The mass shares of isopropyl-alcohol, ethanol and acetone are respectively equal to 95%, 95.1-95.9% and 98.7 % in the vapour mixtures including solvent and water.

The specific evaporation rates for the free surface of three-component mixture “acetone – water - ethyl acetate” are presented in Table 3. The evaporation rate of acetone in three-component mixture is 15 % higher, than its rate in two-component mixture. The overall evaporation rate of three-component mixture including ethyl acetate with high vapour pressure is 1.8 times higher than the rate of two-component mixture “acetone – water”. The mass share of acetone in three-component vapour mixtures is 62 %.

During the storage and the maturation of perfumery and liquid cosmetic products and on other hand the extraction of plant materials by using organic solvents in closed vessels the evaporation of solvents is also observed. The evaporation rates in closed vessels for eight solvents and their mixtures are presented in Table 4. The velocity in airspace over liquid solvent was assumed as zero. The solvent content in the head space of vessel was determined as $x_A \approx \varphi \cdot x_W$, where $\varphi = 0.7$ is the assumed relative saturation of air with solvent vapours. Here, it was taken into account the prolonged storage of solvent mixture in the vessel. The evaporation rate of pure solvents in closed vessel is almost four times less than the rate in case of mechanical ventilation in normal mode (at a velocity of 0.2 m/s). The ration between the evaporation rates in case of ventilated air and still air is equal to 3.7 for two-component mixtures. This relation is 3.5 for the three-component mixture.

Table 2. Specific evaporation rates (kg/(m² h)) of solvents from a liquid film depending on the velocity of surrounding air

Solvent vapours, evaporated from pure liquid solvents or two-component liquid mixtures of solvents	Velocity of surrounding air, m/s				
	0.1	0.2	0.5	1.0	1.5
<i>Liquid film of pure solvents</i>					
Methoxy-propanol	1.2	1.3	1.6	2.0	2.4
Isopropyl alcohol	2.4	2.6	3.1	4.0	4.9
Ethanol	2.6	2.8	3.4	4.3	5.2
Ethyl acetate	8.7	9.3	11.1	14.2	17.2
Hexane	16.8	18.0	21.6	27.5	33.5
Acetone	17.3	18.5	22.2	28.3	34.4
<i>Liquid film of the mixture "ethanol – water"</i>					
Ethanol vapours, evaporated from the mixture	2.68	2.87	3.43	4.38	5.33
Water vapours, evaporated from the mixture	0.14	0.12	0.15	0.19	0.23
Vapours of ethanol and water, evaporated from the mixture	2.82	2.99	3.58	4.57	5.55
The share of ethanol vapours in the mixture, %	95.1	95.9	95.9	95.9	95.9
<i>Liquid film of the mixture "isopropyl alcohol – water"</i>					
Isopropyl-alcohol vapours, evaporated from the mixture	2.50	2.67	3.20	4.09	4.97
Water vapours, evaporated from the mixture	0.13	0.15	0.17	0.21	0.26
Isopropyl-alcohol and water vapours, evaporated from the mixture	2.63	2.82	3.37	4.30	5.23
The share of isopropyl-alcohol vapours in the mixture, %	95.0	95.0	95.0	95.0	95.0
<i>Liquid film of the mixture "acetone – water"</i>					
Acetone vapours, evaporated from the mixture	17.8	19.1	22.9	29.2	35.5
Water vapours, evaporated from the mixture	0.24	0.26	0.31	0.40	0.48
Vapours of acetone and water, evaporated from the mixture	18.1	19.4	23.2	29.6	36.0
The share of acetone vapours in the mixture, %	98.7	98.7	98.7	98.7	98.7

Table 3. Specific evaporation rate (kg/(m² h)) of vapours from a film of three-component mixture of "acetone – water" depending on the velocity of surrounding air

Solvents vapours, evaporated from the liquid mixture	Velocity of surrounding air, m/s				
	0.1	0.2	0.5	1.0	1.5
Acetone vapours, evaporated from the mixture	20.5	22.0	26.3	33.6	40.8
Water vapours, evaporated from the mixture	0.32	0.34	0.41	0.53	0.64
Vapours of ethyl acetate, evaporated from the mixture	12.3	13.1	15.7	20.1	24.4
Vapours of acetone, water and ethyl, evaporated from the liquid mixture	33.1	35.5	42.5	54.2	65.9
The mass share of acetone vapours in the vapours mixture, %	62.0	62.0	62.0	62.0	62.0
The mass share of ethyl-acetate vapours in the vapours mixture, %	37.0	37.0	37.0	37.0	37.0
The mass share of water vapours in the vapours mixture, %	1.0	1.0	1.0	1.0	1.0

Table 4. Specific evaporation rate (kg/(m² h)) from the surface of solvents or their mixtures in a closed vessel

Solvent or mixture	For component 1	For component 2	For component 3	Total
Isopropyl alcohol	0.68	0.68	0.68	0.68
Ethanol	0.73	-	-	0.7
Hexane	4.69	-	-	4.7
Acetone	4.83	-	-	4.8
Isopropyl alcohol - water	0.70	0.12	-	0.8
Ethanol - water	0.75	0.13	-	0.9
Acetone - water	4.98	0.23	-	5.2
Acetone - water - ethyl acetate	5.72	0.30	3.42	9.4

Time for evaporating solvents from a liquid film

These evaluations were done for a film with a thickness 0.2 mm and an area of 1 m². The times for evaporating solvents were calculated taking into account the mass of each solvent in this film and the corresponding its evaporation rate pointed in Table 2. The expecting minimal times for evaporating pure solvents depending on the velocity of surrounding air are presented in Table 5. In practice the evaporation rate is maximal in the beginning and then it declines. The evaporation of these solvents is realised for short time as the latter is in the range of seconds for solvents with vapour pressure more than 10 kPa (see Table 1). In Table 5 the times for evaporating solvents from two-component mixtures are shown as well. The solvents in mixtures are evaporated faster than pure solvents. The more volatile component is evaporated first even when its content is 4-5 times higher in the mixture. The evaporation of water is faster in a mixture with more volatile component as acetone. The expecting times for evaporating solvents from a three-component mixture with volatile acetone are also presented in Table 5. All components even less volatile water are evaporated faster in the range of seconds.

Table 5. Expecting minimal time τ for evaporating the pure solvents from a liquid film with an area of 1 m² and a thickness 0.2 mm depending on the velocity of surrounding air

Pure solvent or mixture	Share, %	Velocity of surrounding air, m/s					Time τ in
		0.1	0.2	0.5	1	1.5	
Pure solvents							
Methoxypropanol	100	9.1	8.5	7.1	5.6	4.6	min
Isopropyl alcohol	100	3.9	3.6	3.0	2.4	1.9	min
Ethanol	100	3.6	3.4	2.8	2.2	1.8	min
Ethyl acetate	100	1.2	1.2	1.0	0.8	0.6	min
Hexane	100	39	36	30	24	19	s
Acetone	100	28	26	22	17	14	s
Mixture "ethanol - water"							
Ethanol in the mixture	75	2.8	2.6	2.2	1.7	1.4	min
Water in the mixture	25	18.0	21.1	16.8	13.3	11.0	min
Mixture "isopropyl alcohol - water"							
Isopropyl alcohol in the mixture	83	3.3	3.1	2.6	2.0	1.6	min
Water in the mixture	17	12.7	11.9	9.9	7.8	6.4	min
Mixture "acetone - water - ethyl acetate"							
Acetone in the mixture	94	26.4	24.7	20.6	16.1	13.3	s
Water in the mixture	4	71.7	67.0	55.9	43.8	36.0	s
Ethyl acetate in the mixture	2	0.9	0.9	0.7	0.6	0.5	s

It was evaluated the time τ^* for evacuating solvent vapours by means ventilating air as well. The results for three pure solvents and their mixtures are shown in Table 6. The masses of solvents evaporated from a surface with area 1 m² are also pointed in this table. It was assumed that the relative saturation of air with solvent vapours is respectively equal to 0 in incoming airflow and 50 % in out-going flow. The results show that the times τ^* for evacuating solvent vapours by ventilation are less the times τ for evaporating the solvents from a free thin liquid film (see Table 5). Therefore the ventilation system (even at normal a velocity of 0.2 m/s) can take off the solvent vapours faster than they can evaporate from a film of liquid spilled.

Table 6. Expecting minimal time τ^* (s) for evacuating solvent vapours depending on the velocity of air ventilated

Solvent	Liquid evaporated	Mass of solvent, g/m ²	Air velocity, m/s		
			0.2	0.5	1.5
Acetone	Pure solvent	156.9	23.1	9.3	3.1
	in a mixture with water and ethyl acetate	150.5	22.1	8.9	3.0
Ethanol	Pure solvent	157.8	143.5	54.4	18.3
	in a mixture with water	126.2	114.7	43.5	14.7
Isopropyl alcohol	Pure solvent	157.2	145.2	58.1	19.4
	in a mixture with water	136.5	126.1	50.4	16.8

Additional discussions

A scenario of emergency spillage was modelled by the latter evaluations for the evaporation time from a thin film of liquid spilled. Under normal ventilation (at a velocity of 0.2 m/s) more volatile pure acetone and hexane from the spot of emergency spill will evaporate up to 40 seconds and less volatile pure ethanol and isopropyl alcohol – for up to 4 minutes. In case of emergency ventilation (at a velocity of 1.5 m/s) more volatile hazard organic solvents (as acetone and hexane) can be evaporated in the range of seconds and less volatile ethanol and isopropyl alcohol – up to 2 minutes. Taking into account the high evaporation rate of hazard organic solvents, it may be recommended in a case of an emergency spill all staff must leave immediately the workroom and press the emergency button for switching on the emergency mode of ventilation. The time evaluated for sucking vapours from a film of liquid spilled is less than five minutes, even when the ventilation works in a normal mode. For that the staff may return in the workroom after ten minutes of an operation of the ventilation system. Let's compare evaporation rates of volatile hazard solvents in case of an emergency spill and a normal mode of operation of liquids with these solvents. The case study is related to a workshop with a capacity 600 L per day (at an 8-hour shift) for producing model mixtures pointed in the section Materials. The solvent losses were assumed as 1.5% per day or 9 L/d and respectively 1.1 L/h liquid cosmetic product.

The corresponding solvent emission will be 0.81 kg/h acetone or 0.74 kg/h isopropyl alcohol, or 0.67 kg/h ethanol. So, the evaporation rate of acetone in case of an emergency spill in an area of 1 m² is 23-42 times higher than this rate during a normal mode of operation. The evaporation rates of isopropyl alcohol or ethanol in case of an emergency spillage is 3.5-6.6 and 4.2-7.8 times higher, respectively.

Conclusions

An analytical approach was developed for evaluating the evaporation rate of solvent from free surface of solvents and their mixtures in case using mechanical ventilation.

The evaporation rates for six volatile solvents, their two- and three-component mixtures were evaluated depending on the velocity of surrounding air. Their specific evaporation rates varies from 1.2 kg/(m².h) (for methoxy-propanol) to 66 kg/(m².h) (for three-component mixture including acetone). In case of an emergency mode of ventilation (at a velocity $v = 1.5$ m/s) the evaporation rate is about two times higher than this rate in case of a normal operation mode (at $v = 0.2$ m/s). The evaporation rate of pure solvents in closed vessel is almost four times less than the rate in case of ventilation in a normal mode (at 0.2 m/s). It was evaluated that the time for evaporating volatile solvents from a liquid film (with a thickness 0.2 mm) vary from 0.5 sec to 9 minutes. The time for evaporating pure solvents with a vapour pressure higher than 10 kPa (as acetone and hexane) is in the range of seconds. The solvent from mixtures is evaporated faster than from a pure solvent.

The results of these evaluations were applied for the design and calculation of ventilation system in the workshops of "Star Nails Bulgaria" Ltd. Plovdiv.

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Note: The present revised version was submitted additionally by the author and takes into account the presentation of all data and results in SI system.