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Pool Boiling of Liquid-Liquid Multiphase Systems

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1. Introduction

Heat transfer to boiling liquid is an important problem in the unit operations of evaporation and distillation and also in other kinds of general processing, such as steam generation, petroleum processing and control of temperatures. In spite of frequently occurring cases of homogeneous liquid boiling, meet quite often in industrial practice the necessity to determine the heat transfer conditions during liquid nonhomogeneous mixture boiling, which the mixtures compose the multiphase systems of mutually insoluble (immiscible) liquids. As example the heating and evaporating processes connected with emulsions and other immiscible liquid systems, such as the water-oil mixtures or oiled refrigerating media, or the thermal processes of coal tar preparation can be enumerated. The last of the above-mentioned examples includes many necessary for tar processing unit operations and also technological processes, such as the tar thermal dewatering and distillation, as well as the watered oil-fraction processing. In each of the mentioned above as well as in many other cases meet the diversified heat transfer terms general for the reason of completely different nature of the liquids, including the physical properties of nonhomogeneous mixture components.

In spite of many theoretical and experimental works the heat transfer conditions - and also the heat transfer coefficient values - for various media are still extremely difficult to determine. There is a lack of general models of boiling process, no agreed views concerning the heat transfer mechanism in such process exist. It will be emphasised, what is quite often noticed in literature (Cieśliński, 1996; Collier, 1981; Hobler, 1986) that the accessible models of pool boiling process permit to determine the heat transfer coefficients only after the suitable constants assuming, as determined on the basis of experimental investigations. It concerns also the pure liquids (or homogeneous mixtures), which the boiling process is relatively well known and described - although the heat transfer terms are being determined first of all on the basis of empirical models with the limited range of application.

A great gap in literature exists with respect to liquid multicomponent mixtures or mixtures consisting of mutually insoluble liquids, while the boiling process of such mixtures is still not mastered enough. The rare works which are considered in literature (Alperi & Mitchell, 1986; Matthew et al., 2009; Mori et al., 1978, 1980) include the cases of durable water-oil emulsion boiling and that only in regard to the situation when the oil is the phase of lower

density than water, also in the situation when a more volatile component of mixture is in direct contact with the heating surface (Gorenflo, 2001). The opposite situation can be met, among others, in cryogenic processes, e.g. during the liquefied gas mixture boiling on the water surface (Boe, 1997), but the peculiarity of such processes does not meet any analogy for the liquid boiling on the heated surface.

In literature can also be found only a few works refer to the structures of boiling liquid-liquid mixtures and its effect on heat transfer conditions. One of these works is Greene's et al., (1998) investigations, relative to the phenomenon of dissipation of the heavier fluid in a lighter as a result of the flow of vapour bubbles. Previously, Mori (1985) described the various configurations of "two-phase bubbles", formed during vapour flow through a system of two liquids, in considering the phenomenon of evaporation and condensation, leading to the formation of an emulsion. A few problems of boiling of emulsions are described (Mori, 1978; Mori, 1980, as cited in Tachibana, 1972 and Satoh, 1973) and also with photographic recording systems phenomena of water-oil boiling (Mori, 1980). The other examples of experimental studies of mixtures boiling of liquid-liquid type are showed in table 1, as cited in literature (Gorenflo et al., 2001).

Generally, the aside from the own investigations, for that so the wide range of proprieties of water-oil mixture components there is the large lack of knowledge in the literature both in the field of description of boiling phenomena and heat transfer conditions. Hence, recognition of kinetics of this process is still insufficient and unsatisfactory, especially for technical raw oil materials. From the very beginning, the source of such juncture leads not only in specific proprieties of the water-oil and oil-water mixtures but as well as in many peculiarities occurring during boiling of these types heterogeneous mixtures. It is necessary to emphasize meaningful stochastic character of this process as a results of development of different structures of the water-oil system as well as – what no concern of homogeneous liquids – significant influence the process time duration on heat transfer mechanism of boiling water-oil mixture at constant heat flux .

The authors considered the problem of mutually insoluble liquid mixtures boiling in the course of testing a refrigeration unit, where boiling of oiled ammonia took place (Witczak, 1993, 1997), as well as during investigations of thermal dewatering of coal tars (Filipczak, 1991, 1993, 1997). In the above mentioned cases, the refrigerating oil and tar liquids were the components whose density was higher than that of the second phase. These components were in direct contact with the heating surface. As they have relatively high initial boiling temperature, the heat transfer conditions are considerably different from those presented in the subject literature.

In other studies it was found (Troniewski, 2001) that the phenomena and peculiarities associated with boiling of such mixtures, resulting from changing of physical properties of mixtures and from heat transfer conditions. This is particularly connected with the structures evolving of liquid-liquid system during the boiling process. It was noted at the same a significant effect of time on development of structures during boiling with constant heat flux, and additionally - depending on the volume fraction of the oil phase in the mixture - the different structures with variable of this heat flux (Filipczak, 2000, 2008; Troniewski 2001, 2003, Witczak, 2008).

In aim of solution to these problems the own investigations was carried out. The paper presents the results of laboratory experiments of pool boiling heat transfer from a horizontal copper plate to the water-oil and oil-water mixtures during nucleate boiling. The purpose of

experiments was to describe the behavior of nonhomogeneous liquid-liquid mixtures during the process of pool boiling and also to determine the conditions of heat transfer. The attempt was made to determine the influence of oil phase content in mixture on the mechanism of boiling and on the value of heat transfer coefficient.

No.	Authors	Heating element	Fluid system	Boiling mode
1.	Bonilla & Perry (1941)	Horizontal heavy chromian plated copper plate	Water/1-butanol	Film and nucleate boiling
2.	Bonilla & Eisenberg (1948)	Horizontal heavy chromian plated copper plate	Water/styrene; 1,3-butadien/water	Film and nucleate boiling
3.	Van Stralen et al., (1956)	Wire	Water/organics	Nucleate boiling
4.	Wastwater & Bragg (1970)	Horizontal plate	R113/water; n-Hexane/water; Water/perchlorethylene	Film boiling

Table 1. Experimental investigation of boiling heat transfer to immiscible liquid-liquid mixtures (as cited in Gorenflo et al., 2001).

2. Laboratory test investigation

General view of experimental facility and devices of experimental set-up is shown in Fig. 1. Research of water and oil mixtures boiling was conducted in a closed cylindrical vessel with flat bottom with a volume about 5 dm³ (Fig. 1a, b). The heating surface of vessel bottom was made of copper with an integrated system of thermocouples and an electric heater with a heating power of 1,2 kW (Fig. 1c). This gave a heat flux $q = 70 \text{ kW/m}^2$. The side wall of the vessel were two, placed opposite each other, sight-glasses, allowing observation and recording video-photo of forming structures. The experimental set-up complements instrumentation and control equipment (Fig. 1d). The pool boiling experiment was conducted for boiling of oil-water mixtures with oils lighter than water (thermal oil of item type), as well as with oil a heavier than water, e.g. anthracene oil-water system. Properties of oils used in the study are given in Table 2 (there are producer operational data). The void fraction of oil in the mixture (ϵ_{ol}) in the case of lighter oils varied in the range of (1÷97)% vol, and anthracene oil – at (5÷30)% vol. At the beginning of each experiment, care was taken to obtain two liquids in form of clearly stratified system. The mixture was brought to a boil and then conducted research in two ways. The first was performed by a series of measurements, at increment of heat flux until it reaches its maximum value. The second way - it was carrying out the process of boiling for a long time at a constant heat flux. The measuring system is provided with a condenser, where the vapor condensed and recycled them into the vessel, thus ensuring the stability of the mixture composition. At the test time, regardless of the observation and recording of structures, it was measured the bottom and bulk temperature, which allowed to determine the conditions of heat transfer.

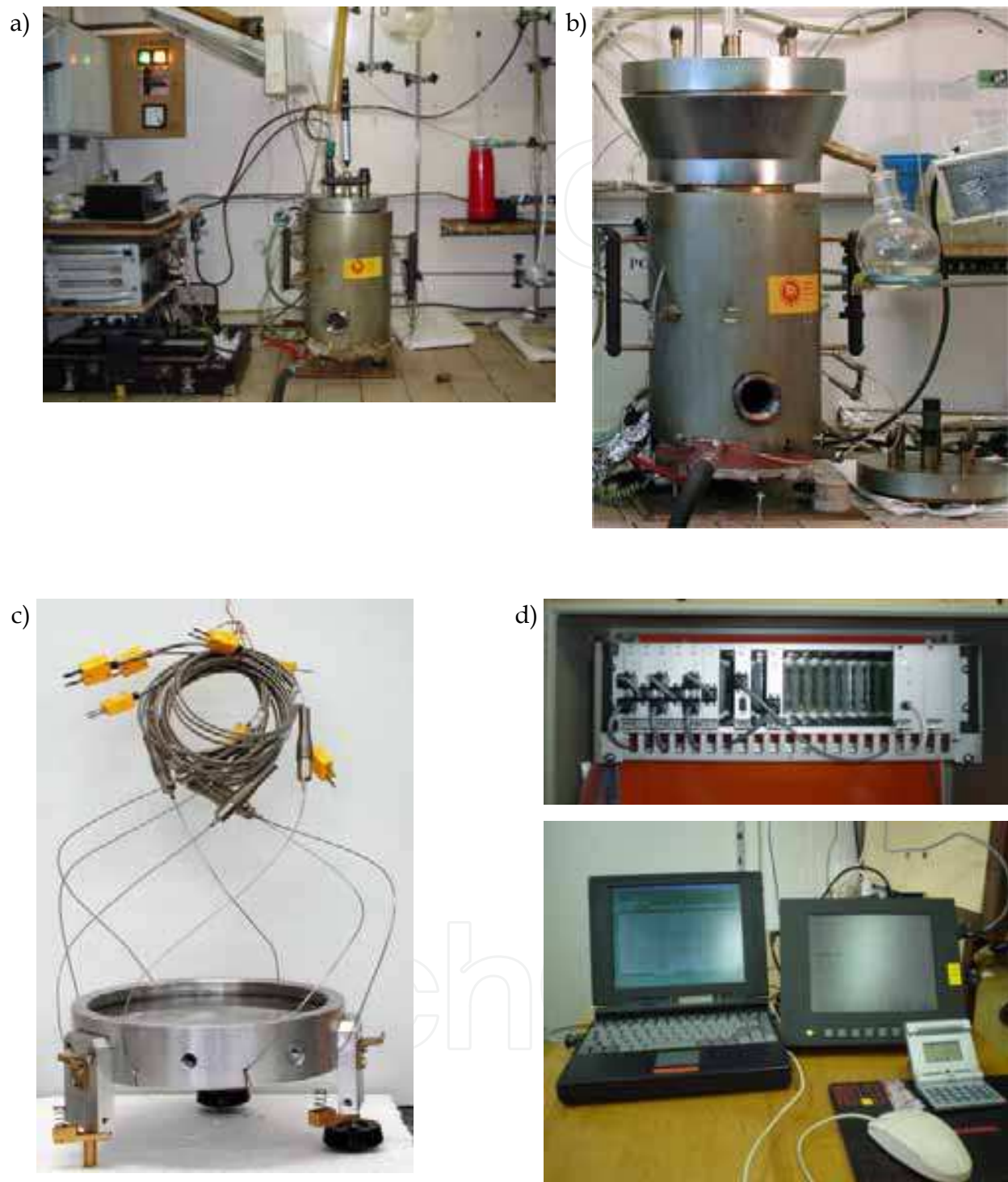


Fig. 1. Heat transfer apparatus: a) general view of experimental facility; b) vessel with top vapour condenser; c) heating plate with bottom thermocouples; d) devices of experimental set-up.

Type of oil	Density, ρ kg/m ³	Viscosity, $\eta \cdot 10^3$ Pa·s	Surface tension, $\sigma \cdot 10^4$ N/m	Boiling temperature, t_b °C
Mineral oil: Iterm oil 6Mb	$\rho_{100} = 820,7$	$\eta_{100} = 3,9$	$\sigma_{99} = 153,2$	347
	$\rho_{26,5} = 863,7$	$\eta_{25,0} = 69,0$		
Mineral oil: Iterm oil 12	$\rho_{100} = 834,8$	$\eta_{100} = 10,3$	$\sigma_{99} = 214,1$	340
	$\rho_{26,5} = 878,6$	$\eta_{25,0} = 360,3$		
Mineral oil: Iterm oil 30 MF	$\rho_{100} = 853,5$	$\eta_{100} = 24,5$	$\sigma_{99} = 241,1$	353
	$\rho_{26,5} = 895,9$	$\eta_{26,5} = 1157,4$		
Anthracene oil (tar oil)	$\rho_{99} = 1088,5$	$\eta_{97,0} = 2,6$	$\sigma_{99} = 266,0$	350
	$\rho_{20,0} = 1129,9$	$\eta_{26,5} = 58,5$		

Table 2. Primary properties of oils used in the experiments.

3. Bulk structures and boiling patterns

According to the data on pool boiling process of immiscible liquids mixtures changing conditions of heat exchange are associated by different in nature characteristics and process parameters.

For example, during bulk boiling of oily refrigerants (Greene et al., 1988), changing the conditions of heat transfer usually occurs as a result of foaming. This effect is due to motion of vapor bubbles through the interfacial area of two immiscible components. In demonstrative way, this mechanism is shown in Fig. 2. As is clear from the presented sketch, the movement of the bubbles gives rise to turbulence in both phases of the liquid, leading to their dispersion.

If the number of bubbles entering the contact area of both liquid-liquid layer is large enough, as a result of intensive mixing may cause to produce the emulsion with different degree of stability.

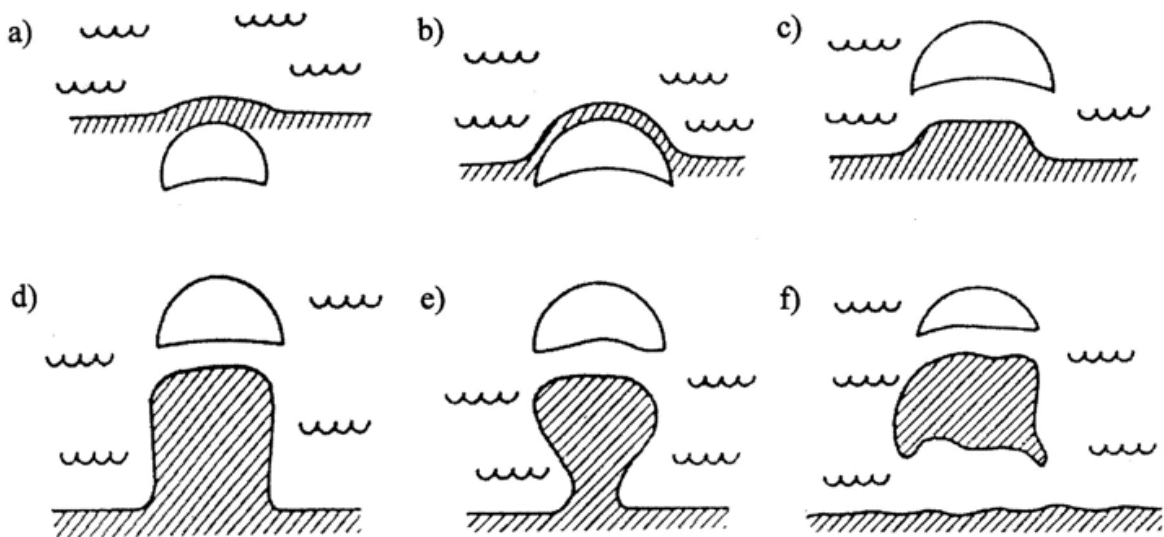


Fig. 2. Behavior of a vapour bubble (stages a÷f) upon boiling of an immiscible liquids mixture (Greene et al., 1988).

The much more complex structures occur during boiling watered coal tars or mixtures of water and oil (with presence of oil lighter and heavier than water) which has been described, at times detailed (Alperi R. W. & Mitchell R.D., 1986; Filipczak G., 1997, 2008; Troniewski L. et al., 2001, 2003; Witczak S. et al., 2008). The result of investigations show that in all these cases the forming structures can to have both the nature of the system with varying extent of stratification of the liquids, and a high area of dispersion or dissipation, until to the emulsion form with different durability as a quasi-homogeneous systems.

3.1 Structures description

The experimental study showed that independently by the type of water/oil/water mixture in the bulk boiling process are observed the same in the picture the specific stages (phases) of the process, which can be grouped as likewise in their form, as it shown in Table 3.

The images presented in the table show that for all tested non-homogeneous mixtures, there is a similar systematics of changes in the structure during the bulk boiling (time factor). There is also any intermediate structures, what is however difficult to interpret, due to the strong dynamics of the boiling process. Attention should be also paid to the fact that many of the same or similar structures - especially of an emulsion or foam - are formed spontaneously, on different values of heat flux. Formed elsewhere in this way structures may be the same more or less stable, which is realized in the quantitative composition of the mixture as well as the duration of the process.

In general heterogeneous structures (NJ) are presented, for which ones we can distinguish a continuous phase (oil or water) and quasi-homogeneous structures (QJ). In the case of heterogeneous structures the individual components of the mixture can be easily distinguished. For the structures of quasi-homogeneous visual distinction of the individual components is very difficult or even impossible. However, in both cases the type of mixture must be recognized due to a density relationship between each one.

As we see from table 3, the systematics of structures is grouped as follows:

1. water-oil system (OLW), where the water is continuous phase, and oil void fraction is $\varepsilon_{oi} \leq 50\%$ vol;
2. oil-water system (WOL), where the oil is continuous phase, and void fraction not exceed 50%;
3. water-anthracene oil system (OCW), i.g. mixture with participation of oil with higher density than density of water - for this system both phases can be continuous.

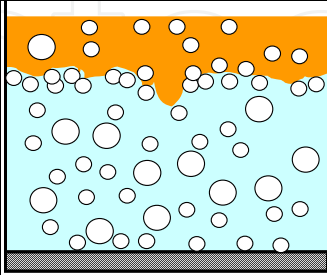
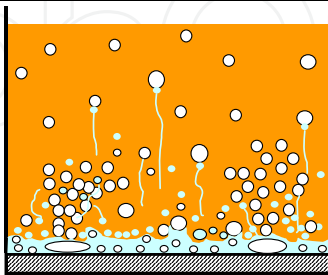
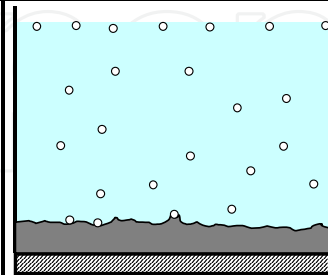
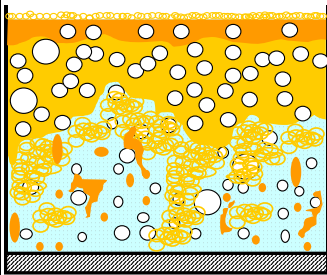
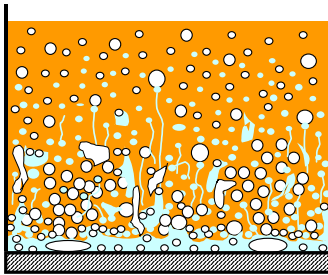
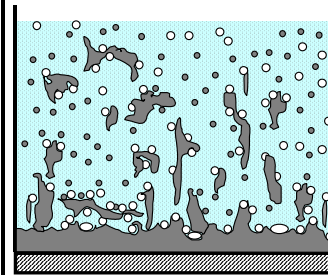
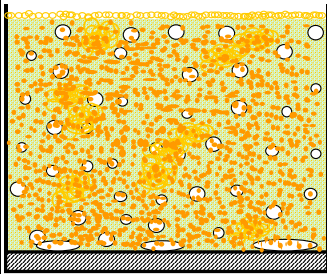
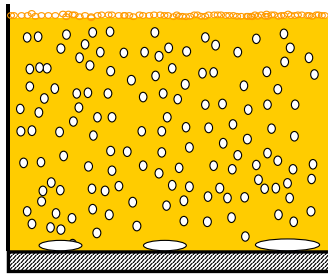
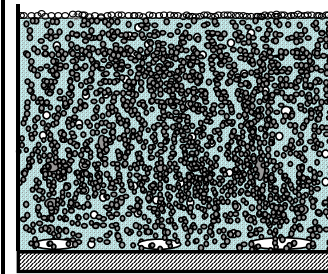
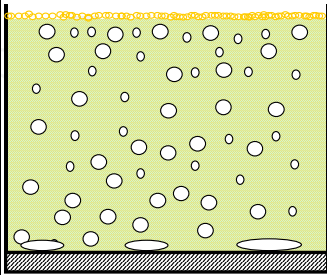
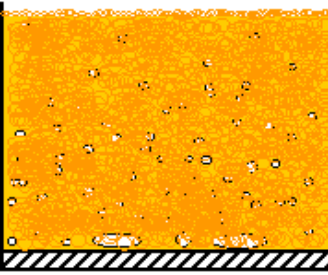
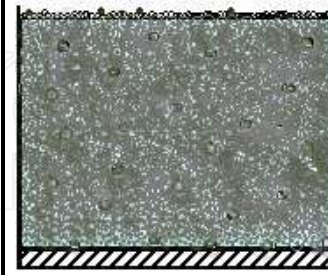
According to phenomenological view for all non-homogeneous mixtures the similar structures were observed: from stratified (a), through stratified-droplets (b), to droplets emulsion or foam (c) and permanent oil/water emulsion or dynamic foam (d).

The real views of identified structures are shown in Fig. 3 to Fig. 5. There are presented the structures for water and oil mixtures (WOL/OLW) with the oil density lower than density of water (Fig. 3 and Fig 4), and the anthracene oil-water mixtures (OCW) with density of oil higher than water density (Fig. 5), respectively.

For pool boiling of liquid-liquid mixtures the systematic of structures is described as follows:

R- stratified structure; RK- stratified-droplet; RKE- stratified-droplets with emulsion; EK- dynamic droplet emulsion; E- dynamic or stability emulsion in the bulk; REP- stratified emulsion with dynamic foam; P- foam in the whole bulk.

Each detailed description of structure is described by dual names, separated by a hyphen (a kind of mixture/structure), e.g.: WOL-P - water/oil - dynamic foam structure (P) in the bulk, OCW-RK - oil/water stratified-droplet structure (RK).

Description	Kind of liquid-liquid system (continuous phase on the first place)		
	water-mineral oil (OLW)	mineral oil-water (WOL)	water-tar oil (OCW)
	$\rho_{ol} < \rho_w$		$\rho_{ol} > \rho_w$
heterogeneous patterns image (NJ)			
a) stratification rippled by vapour bubbles			
b) oil droplets and emulsion areas rushing			
quasi-homogeneous patterns image (QJ)			
c) labile emulsion or emulsion breaking			
d) fully durable emulsion			

← heat flux increasing

heat flux increasing
←

Table 3. Structures and phase transitions at boiling of liquid-liquid mixtures: a) separated structure (R) - sloshing of boundary interface; b) stratified-oil-droplet (RK/RKE) - separated-drops regime (breaking of oil phase and forming of oil droplets with partial emulsification and disintegration of oil droplets oriented on formation of water-oil emulsion); c) oil-droplet emulsion (EK), eventually foam (P) – dynamic droplet emulsion (dissipation of oil droplets); d) permanent emulsion (E) – liquid-liquid emulsion (progress of oil droplets dissipation and formation of a stable emulsion)

Sometimes these changes occur very suddenly. It is a characteristic symptom for so called flash (explosive) boiling, observed practically for all investigated water/oil/water systems. In many cases this leads to the formation of very stable emulsions, what is characteristics for non-Newtonian fluids, with kinematic viscosity several times (one thousand and more) higher than the viscosity of the liquids forming a two-phase liquid-liquid system.

Furthermore, in case of anthracene oil-water mixture boiling (OCW), the emulsion creation phenomenon takes place at the lower value of heat flux density, than in case of water-oil mixture. The state of mixture foaming appears more quickly, as well. For the obvious reasons (lack of liquid transparency) the precise observation of the phenomenon was impossible in this case. Moreover, the high rate of water steam bubbles created results in the high dispersion of oil phase, particularly in the vicinity of heating plate, where the sudden emulsion formation takes place. The emulsification intensifies at the further increase of heat flux density, what leads to mixture foaming till the "boiling over" effect is obtained.

This is important that appearance of each structure can create with increasing or at constant value of heat flux. In the second case ($q=const.$) the time factor is very important - we distinguish the different transition time from stratified forms to more or less homogeneous.

From phenomenological point of view the structures of the boiling liquid-liquid mixtures are as follows:

1. **Water-oil mixtures (OLW) – Fig. 3 - heterogeneous structures (NJ):**

OLW-R - stratified structure. Oil layer located on the water is broken down by the vapour bubbles climbing to the top. The wavy motion is observed after initially stable interphases boundary;

OLW-RK – stratified-droplet structure. The dissipation of mixture components take place as a result of turbulence in the liquid, formed by vapour bubbles move to the top. From the oil layer are pulled his portion in the form of drops of different size and shape and accumulate down to the water volume. On the other hand portions of water are entrained into the oil layer. As a consequence the foam is formed as a thin layer, floating on the free surface of the mixture in the form of large floccules, which tend to hold just below the surface layer oil. As a result of turbulence in the mixture, in great number portions of the oil phase are entrained in the heating plate area;

OLW-RKE – stratified-droplets with emulsion. Non-transparent layer of emulsion is formed, which expands in the direction of the heating plate. A significant reduction in transparency of the mixture due to the presence of oil follows in the form of droplets, a portion of emulsions and foam. The oil in increasing amounts is deposited on the surface heating.

2. **Water-oil mixtures (OLW) – Fig. 3 - quasi-homogeneous structures (QJ):**

OLW-EK – dynamic droplet emulsion. The oil is dispersed as small droplets, and different clouds of foam, circulating in the volume of the mixture. The structure is periodic, partial stratification, becoming more transparent. Structure is accompanied by occasional flash evaporation;

OLW-E – dynamic or stability emulsion. As a result of further dissipation of oil phase the non-transparent emulsion is formed in whole volume of the mixture. Boiling is accompanied by explosiv (flash) evaporation of emulsion.

3. **Oil-water mixtures (WOL) – Fig. 4 - heterogeneous structures (NJ):**

WOL-R - stratified structure. Oil layer forms remains on a layer of boiling water. Periodic structure is accompanied by flash evaporation and release of a portion of steam and water droplets to the volume of oil. As a result a sudden breaking initially stable phase boundary take place;

WOL-RK – stratified-droplet structure. Vapours bubbles are rising into the top layer of oil and leave behind traces of water (micro-droplets of water). At the interface exists a layer consisting of vapour bubbles, drops of water and oil. As a result of flash (explosive) evaporation components of mixture are mixed intensively. The portions of oil are deposited on the heating plate under the influence of turbulence. Further dispersion of phases leads to emulsion in the bulk volume (WOL-E);

WOL-REP – stratified emulsion with dynamic foam. On the emulsion layer remains a dynamic layer of foam formed by the foaming of the emulsion (WOL-E). There has been a rapid increase of the mixture volume and sudden flash evaporation. The structure may take in the emulsion form in whole volume (WOL-E) or the dynamic foam in the bulk volume (WOL-P).

4. **Oil-water mixtures (WOL) – Fig. 4 - quasi-homogeneous structures (QJ):**

WOL-E – dynamic or stability emulsion. The structure is almost totally non-transparent. It is accompanied by explosion evaporation at the beginning of formation of structures very rapidly, and then its intensity decreases. There are boiling with formation of quite a number of small bubbles. The structure may evolve in the stratified emulsion and foam (WOL-REP), and next to foam in the whole volume (WOL-P).

WOL-P – dynamic foam structure. The mixture is totally non-transparent. It is characterized by sudden increase volume of foam and periodic stratification to emulsion-foam structure (WOL-REP).

5. **Water-anthracene oil system (OCW) – Fig 5 – heterogeneous structures (NJ),**

OCW-R – stratified structure. Both phases are clearly separated. Steam bubbles are formed on the layer of oil, which is behind on the hot wall of the heating plate (the oil temperature is above the saturation temperature of water). The movement of vapour bubbles causes the oil layer surface wavy motion.

OCW-RK – stratified-droplet structure. Forming and moving upward vapor bubbles cause turbulence in the mixture, which kidnap oil from the surface layer. Oil droplets are formed with extended shapes. Boiling takes place on the much overheated portions of oil. Further dissipation of oil in water volume leads to decreases thickness of the oil and its breakdown take place.

6. **Water-anthracene oil system (OCW) – Fig 5 – quasi-homogeneous structures (QJ):**

OCW-EK – dynamic droplet emulsion. The emulsion takes the form of small oil droplets dispersed in water volume. As a result of tearing of oil from bottom the water evaporates very rapidly by contact with preheated plate. There is flash (explosive) evaporation and intensive dissipation of oil. Depending on heat flux a periodic stratified-droplet (OCW-RK) or dynamic foam (OCW-P) are formed.

OCW-P – dynamic foam structure. Consequently, by flash evaporation of water produce non-transparent dynamic foam in the bulk mixture. This causes a rapid increase of mixture volume.

It should be noted that at boiling of the water-anthracene oil (Fig. 5) we have different, in relation to other tested mixtures, mechanism of formation of structures. This follows from the fact that anthracene oil is keeping on the heating plate, what delays the process of boiling water. The same time with the increase of heat flux leads to changes of structures of this system what is very intense, because they occur as a result of flash evaporation of water after contact with a highly superheated wall of heating plate. Additionally, in this case do not exist the conditions conducive to the formation of more stable emulsion, and only highly dynamic foam (totally non-transparent), which is usually re-stratification.

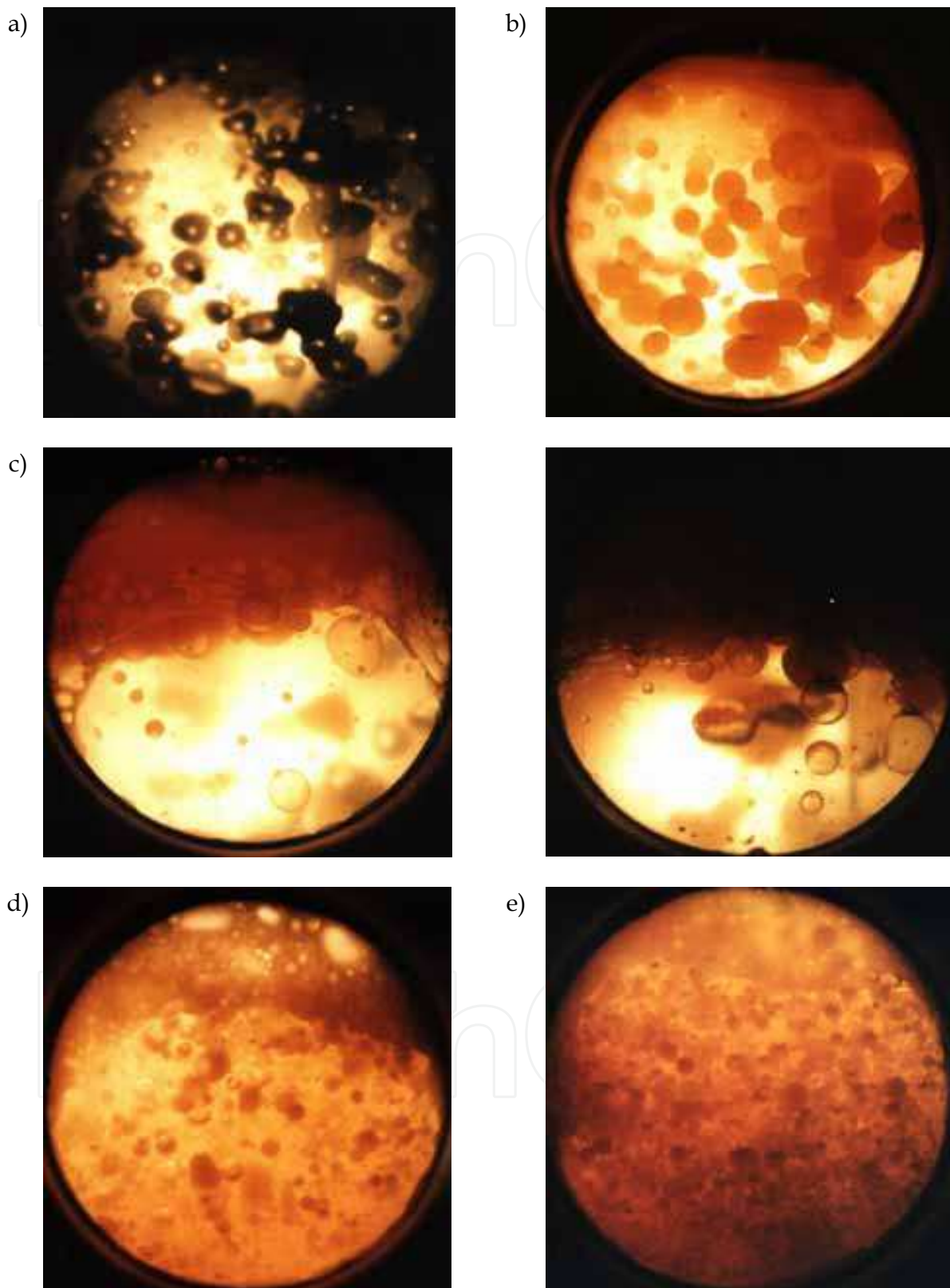


Fig. 3. Boiling structures of water-oil mixture (OLW). Nonhomogeneous structures (NJ): a) OLW-R, b) OLW-RK, c) OLW-RKE; Quasi-homogeneous structures (QJ): d) OLW-EK, e) OLW-E

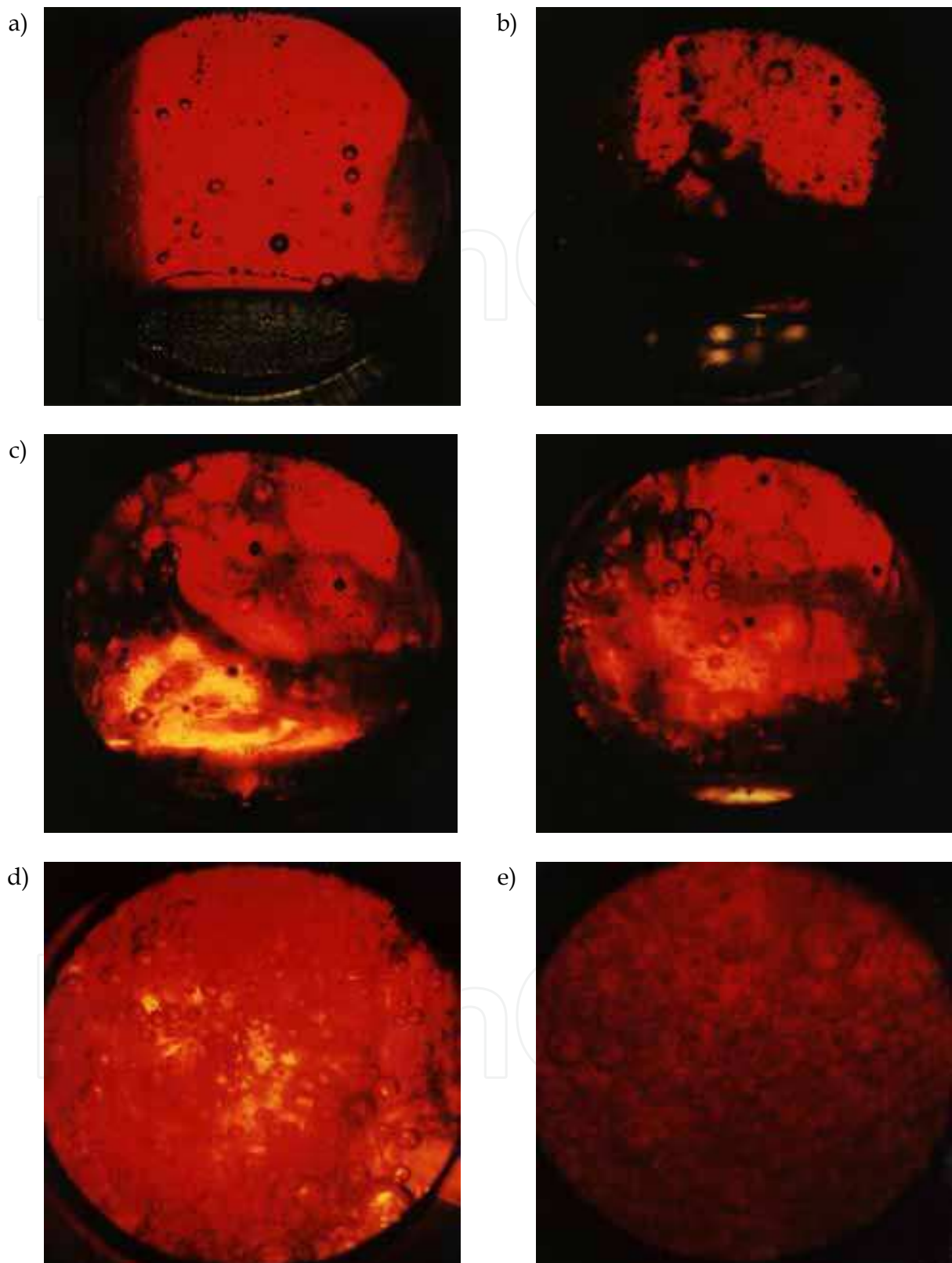


Fig. 4. Boiling structures of oil-water mixture (WOL). Nonhomogeneous structures (NJ): a) WOL-R, b) WOL-RK, c) WOL-REP; Quasi-homogeneous structures (QJ): d) WOL-E, e) WOL-P

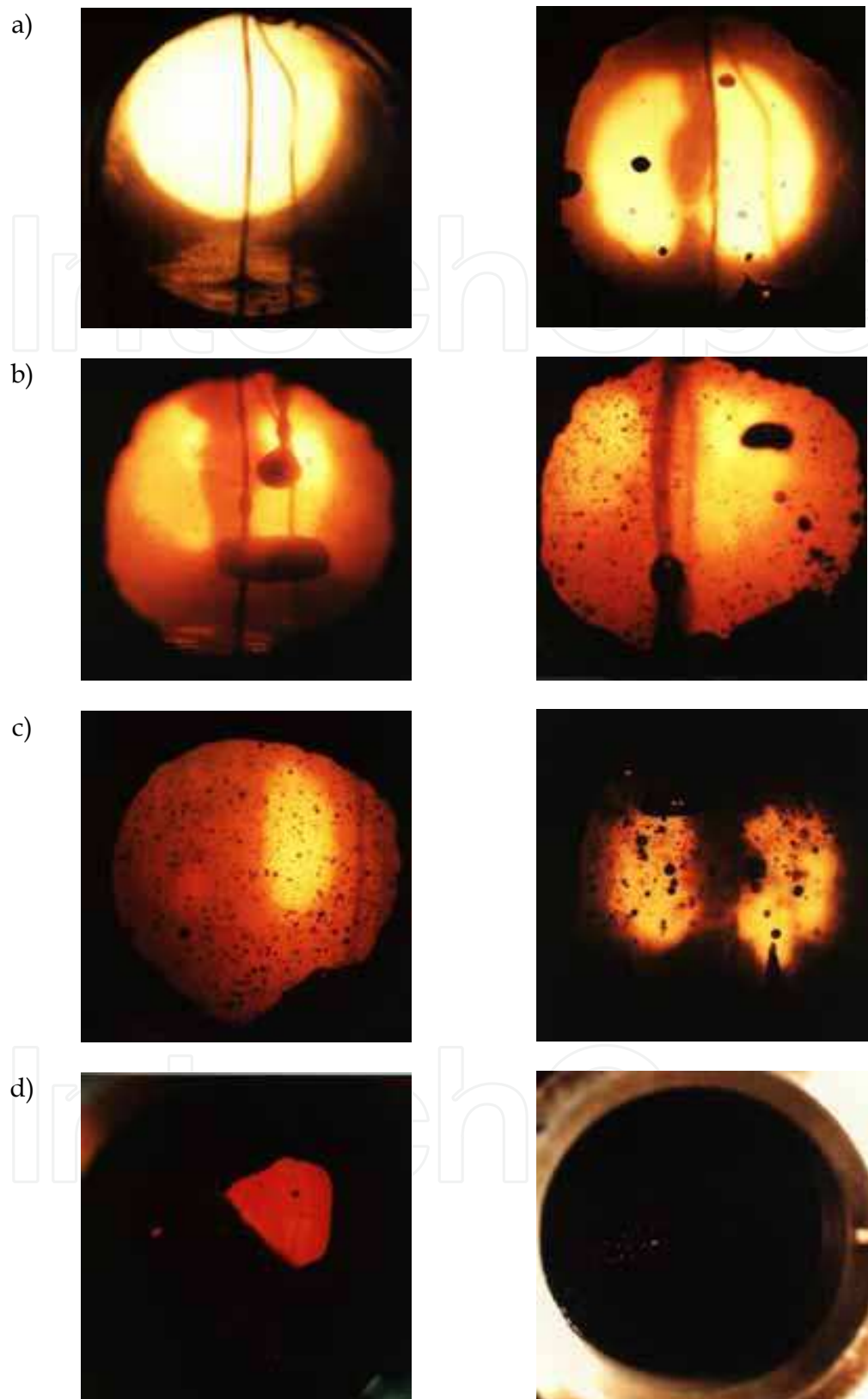


Fig. 5. Boiling structures of water-anthracene oil mixture (OCW). Nonhomogeneous structures (NJ): a) OCW-R, b) OCW-RK; Quasi-homogeneous structures (QJ): c) OCW-EK, d) OCW-P

To illustrate of effect resulting from the dynamics of the process in Fig. 6, for example, shows photographs of two-phase system water/oil/water obtained as a final result of the boiling process in the same conditions, but at different times (sometimes from technical reasons it was impossible to recording changes in the structure of the boiling). Studies have found out that, according to the type and composition of the mixture and heat flux, observed strongly different rates the transition from heterogeneous stratified mixtures to quasi-homogeneous emulsion and foam.

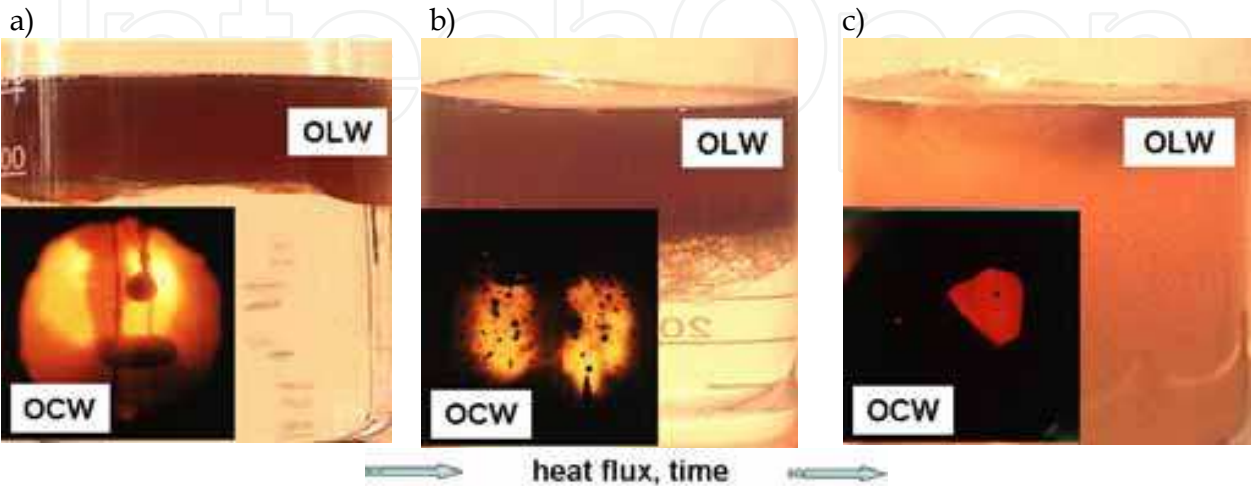


Fig. 6. Comparison of phenomena boiling of oils with different density than water (OLW/OCW) versus heat flux and time: a) separated oil-water system; b) water-emulsion-oil mixture; c) emulsion in whole volume of liquid-liquid mixture

Create of different structures were observed both at increase and fixed in time the heat flux density. It was found that type of formed structure first of all depends on the heat flux density and duration of the process and the volume oil fraction and a small extent of its kind. Heterogeneous structure occurred first of all in the initial period of boiling to smaller values of heat flux, a quasi-homogeneous structure of the average and larger values of this one. Emulsions obtained as a result of boiling were characterized by different stability - the more oil in the mixture, the more stable emulsions. Some of them showed very low stability (dynamic emulsions) and almost immediately after completion of heating was observed stratification - especially at low contents of oil phase. In the other cases emulsions are breaking after a few hours or not at all (permanent emulsions). Should be noted that these emulsions showed a characteristics of non-Newtonian shear thinning fluid, with viscosity of many times more than the viscosity of the used oils (in ambient temperature). The images of microstructures, for example, for the three produced during boiling of emulsions are shown in Fig. 7.

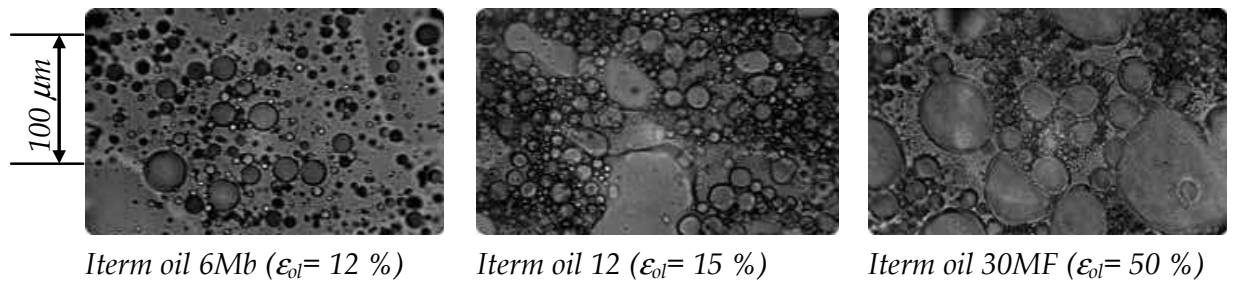


Fig. 7. Microstructures of water-oil emulsions obtained during boiling.

3.2 Dissipation of water-oil systems

As a result of observation of boiling mixtures, it was found that the mechanism dissipation of the components during the movement of bubbles through a layer of heavier and lighter component is more complex than that presented in literature. For mixtures type WOL/OLW were observed first of all the oil dissipation in water. For more volume fraction of oil increased the oil dispersed in water. In this case the water is transported to the volume of oil as a result of turbulence of steam bubbles. This mechanism is analogous to that shown in Fig. 2, as well as in the work of Greene et al., (1998). However, for boiling oil-water mixture the temperature of oil phase is always slightly lower than the temperature of boiling water and steam bubbles condense and takes the form of two-phase bubbles, what is clearly shown in Fig. 8.

Dissipation of micro-droplets of water takes the form of traces of water (water-marks) causing clouding oil (Fig. 8a). As a result of intense evaporation to colder oil get into the larger water droplets formed from condensation of large bubbles of steam (Fig. 8b). It was also found the presence of two-phase bubbles in the form of one or two bubbles trapped in the upper part of a drop of water (Fig. 8c).

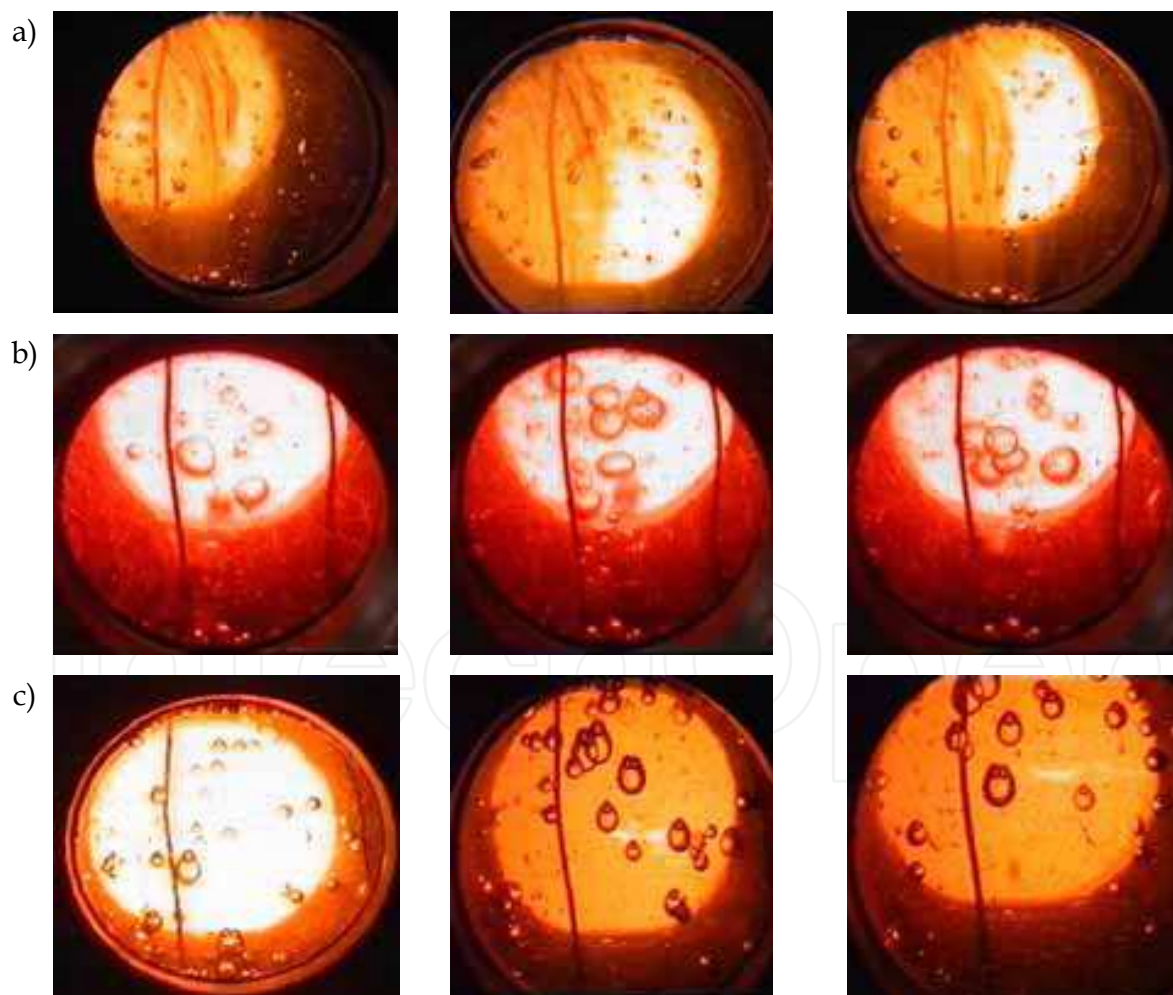


Fig. 8. Configurations of two-phase bubbles in water-oil mixtures ($\varepsilon_{ol} = 76\%$): a) water-marks; b) big steam bubbles and water drops; c) two-phase bubbles

For oil-water mixture with anthracene oil (OCW) the void fraction of oil did not exceed 30%. In this case were observed first of all dissipation drops of oil in water volume – see Fig. 5b.

As a result of turbulence arising from the movement of vapor bubbles upward portions of oil are extracted to a volume of water from the oil layer, deposited on the heating plate. The intensity of dissipation of oil significantly increases when the water after breaking the oil layer penetrate the plate directly. As consequence the rapid evaporation and intense mixing of the components of the mixture giving rise to unstable emulsions and dynamic foam. Detailed analysis of this phenomenon has proved be impossible because of the loss very quickly transparency.

4. Heat transfer conditions - results of investigation

On the terms of heat transfer during boiling of mixtures of immiscible liquids is highly influenced by different physical and chemical properties of mixture components and hydrodynamic phenomena associated with boiling (cross dissipation of water and oil phase inversion). These phenomena - both dependent on the proportion components and their properties - make the process of boiling the liquid-liquid mixtures differs significantly from boiling of pure components. These differences manifest themselves primarily through the formation of different of structures that determine the conditions of a significant heat transfer. It should clearly emphasized that the literature is not encountered as an accurate and complete systematics of structures formed during boiling liquid systems mutually immiscible.

Carried out own research confirmed the significant complexity of boiling water and oil mixtures, and the explicitly impact of structures formed on the conditions of heat transfer. The characteristic feature of the process is the phenomenon of flash (explosive) evaporation occurring practically for every examined system, even with a small amount of oil in the liquid-liquid mixture. This involves sudden formation of large bubbles of steam, with even a few centimeters in diameter. This was accompanied by a loud crash, launch large amounts of the mixture and sudden upward mixing of components as a result of the creation powerful turbulence in the mixture.

The typical investigation results covered comparison the values of heat transfer coefficient for boiling of water as well as a free convection for examined water-oil systems are presented in Fig. 9. The experimental data indicate that for the same heat flux density (q), the values of heat transfer coefficient for pure water boiling are average several times larger than for the conditions of heat transfer with oil phase. The significant peculiar occurring during boiling of water-oil mixtures is decrease in efficiency in heat transfer called in the literature as damping effect. The scale-size of this effect depends on the time factor of process as well as on the mixture structures – Fig. 10 and Fig. 11.

Independently from contribution of phases and heat flux, the formation of a specific structure which determines both the heat transfer conditions always requires a period of time. This is connected with ever-changing structures of liquid-liquid, which very often is not permanent and appear alternately and randomly.

The aim of illustrating influence a detailed nature of the boiling time for the test mixtures in Fig. 12 to Fig. 14 illustrated the duration of the process in relation to temperature ΔT , denoted as the difference between the wall temperature of heating plate and saturation temperature of water. Additionally, the images of structures which were registered at the time of boiling process are shown.

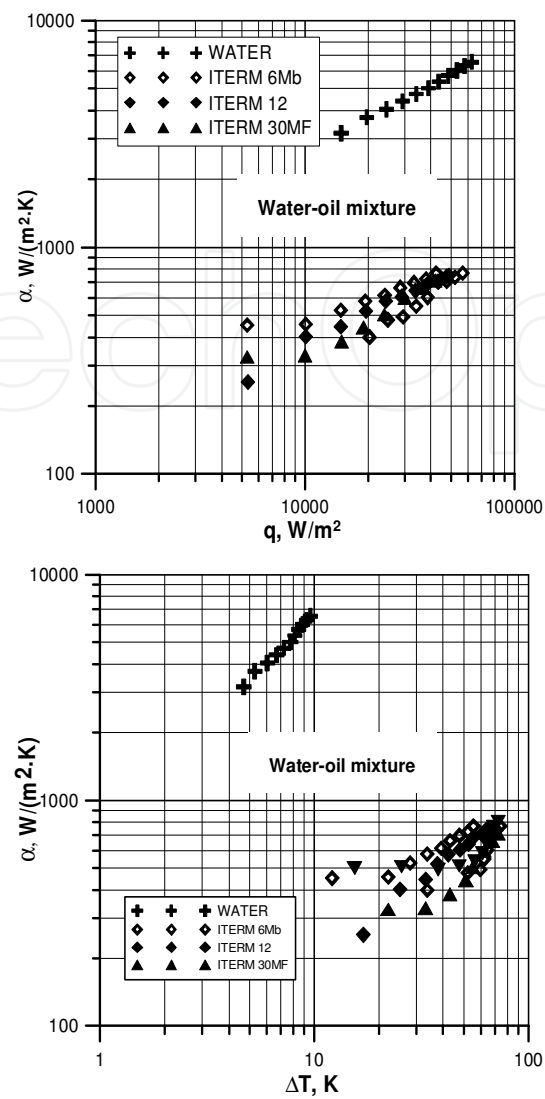


Fig. 9. Heat transfer coefficient for water and water-iterm oil mixtures

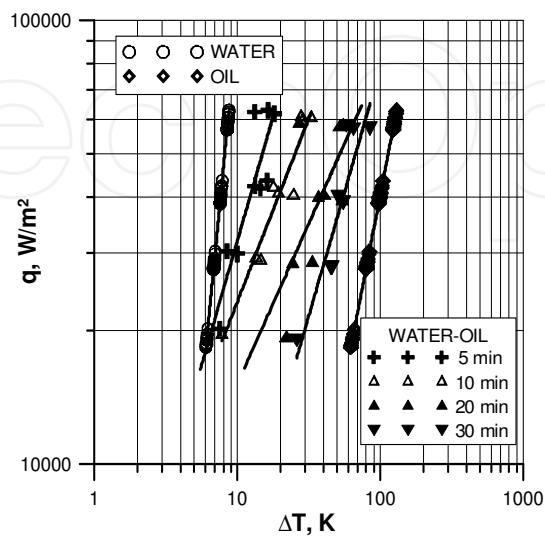


Fig. 10. Time-boiling curves for water-oil item 12 mixture, ($q=18\div67,8\text{ kW/m}^2$, $\varepsilon_{ol}=12\%$ vol.)

The experimental results show a clear increase in temperature difference, from a few to several dozen degrees. The characteristic feature is fact that this difference increases monotonically, which indicates a uniform heating of the system, regardless of the type of structure. It should be noted that the presented results refer to the average temperature of the wall, so the drawings are not visible to the local (instantaneous) changes in temperature. Consequence of changes of temperature difference ΔT is changes of heat transfer coefficient, which illustrated in Fig. 15.

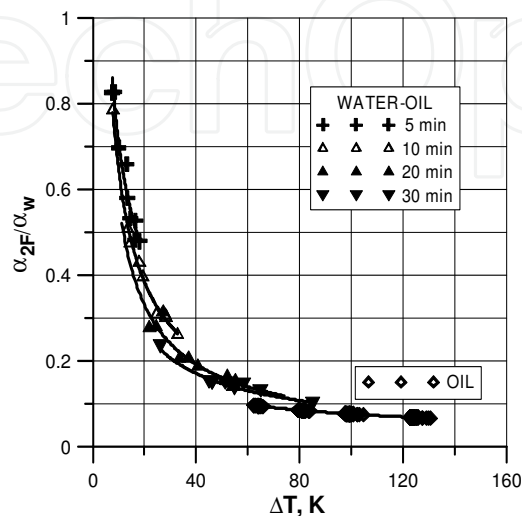


Fig. 11. Boiling damping effect for water-oil item 12 mixture ($q=18\div 67,8 \text{ kW/m}^2$, $\varepsilon_{ol}=12\% \text{ vol.}$)

The experimental results show very significant correlation between the time-scale and the water-oil structures forming during of boiling process. Moreover, data experiments shown in these figures indicate that in the range of so-called heterogeneous structures – Fig. 12b, Fig. 13b and Fig. 14b – the boiling process has non-steady character, whereas for quasi-homogenous ones which staying more or less permanent water-oil emulsions in the bulk mixture, the process has steady or near steady attribute.

Than, for heat transfer coefficients relation α_{2F}/α_w , versus temperature difference ΔT (Fig. 14), we can distinguish two typical of nature of the process regions in which the water or the oil are predominant phase, respectively, for heat transfer mechanism and which are characteristic of damping effect (cp. Fig. 11).

It can be observed that the greatest heat transfer coefficient values are in the delaminated of structures (OLW-R, OLW-KR), when the progressive with the duration of the process of change to achieve an order of lower values for dynamic of structures quasi-homogeneous (OLW-EK, OLW-E). This trend does not depend from the type of oil, but only the duration of the process, with whom the change of structure is associated.

Experimental results and observation of process phenomena convince about very important influence of the process run-time on boiling mechanism. As an example, in Fig. 16 the course of events versus time including heat transfer coefficient (α_{2F}) are presented, bulk mean temperature distributions (T_m) and difference of temperature between the heat surface and saturation temperature of water (ΔT_m), all at 12% vol. of oil void fraction. We see sudden change of the values of these parameters and in consequence heat transfer coefficient redistribution, particularly for separated and heterogeneous ranges of mixture structures. Past a certain limiting time (τ_{lim}), in the consequence of forming quasi-homogenous structures, heat transfer conditions staying more stabilized and that situation remains unchanged still the long time.

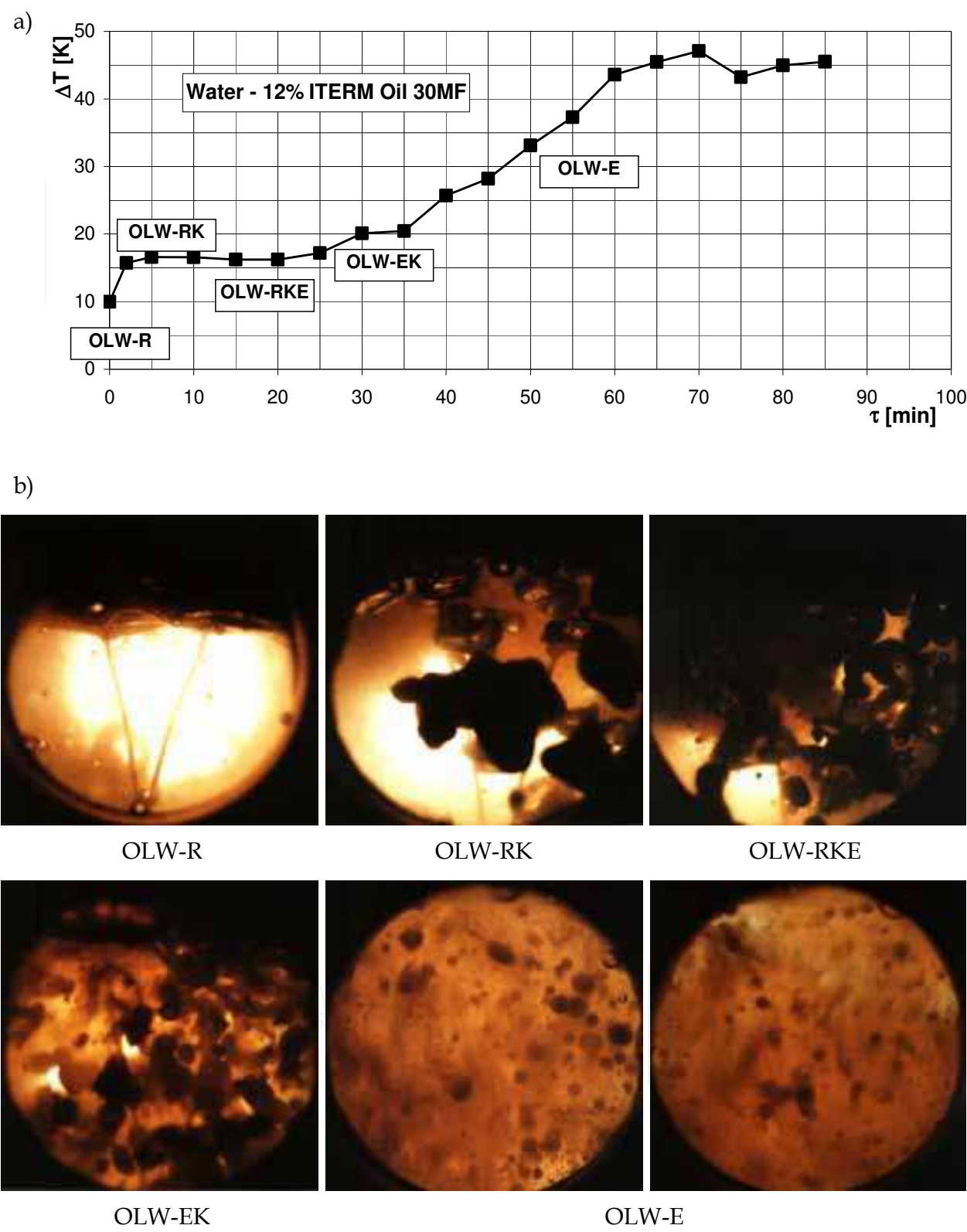


Fig. 12. View boiling run-time of water-oil item 12 mixtures: a) temperature difference; b) image structures – heterogeneous (R/RK/RKE) and quasi-homogenous (EK/E); ϵ_{ol} =12% vol., q =43,5 kW/m²

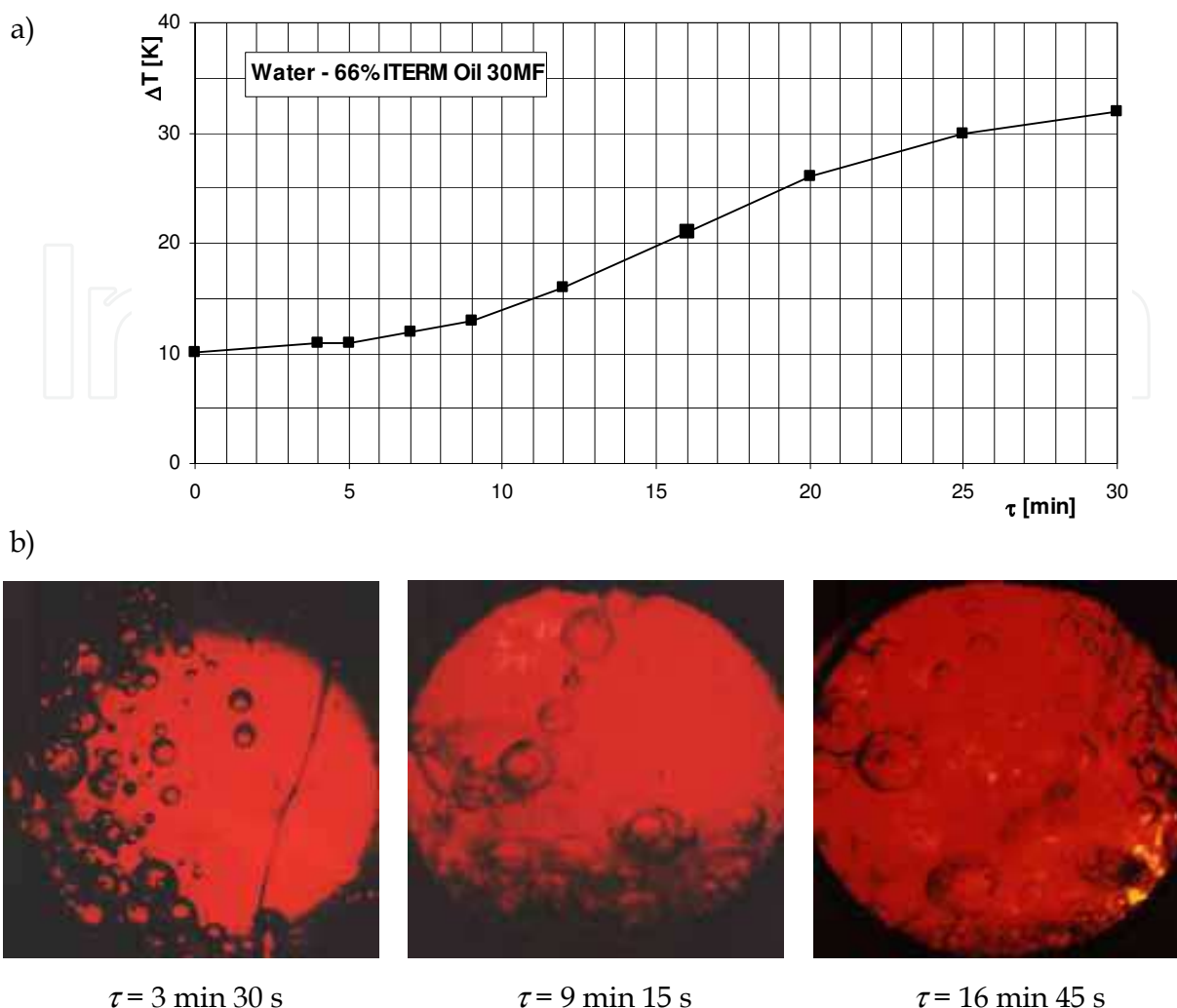


Fig. 13. View boiling run-time of water-oil item 12 mixtures: a) temperature difference; b) image structures ($\varepsilon_{ol}=66\%$, $q=19,5 \text{ kW/m}^2$)

The explicit characteristic of process is to make equal after a time more than the time limit ($\tau < \tau_{lim}$) the value of this ratio with the rise of quasi-homogeneous structures. This reflects a same time developing the mechanism of heat transfer in this type of mixtures (in the studied range of process parameters). Nevertheless, the results shown in Fig. 16 and Fig. 17 indicate that on this mechanism also has influence the surface power heat. This can be seen by changing the shape of the boiling curves for different values of heat flux what indicates its significant influence on the conditions and formation of the structures of boiling water-oil mixture.

Take into consideration such mechanism for different water-oil mixtures – Fig. 17 – we come to a conclusion that quantitative solutions of boiling process should refers to inversion phase of components of oil-water mixture. Until now this variability was explained by structures of liquid-liquid mixtures and by their form heat transfer conditions was denoted. However, the study indicate that a decisive impact on this state of affairs is that the concentration of oil phase (ε_{ol}^*), as well as stable oil-water emulsion, on the surface heating (Filipczak, 2008; Troniewski, 2001; Witczak, 2008). The results show that a small amount of oil already in (on) the surface heating, appearing also in the stratified structure (Tab. 3), leads to very large disturbance in heat transfer.

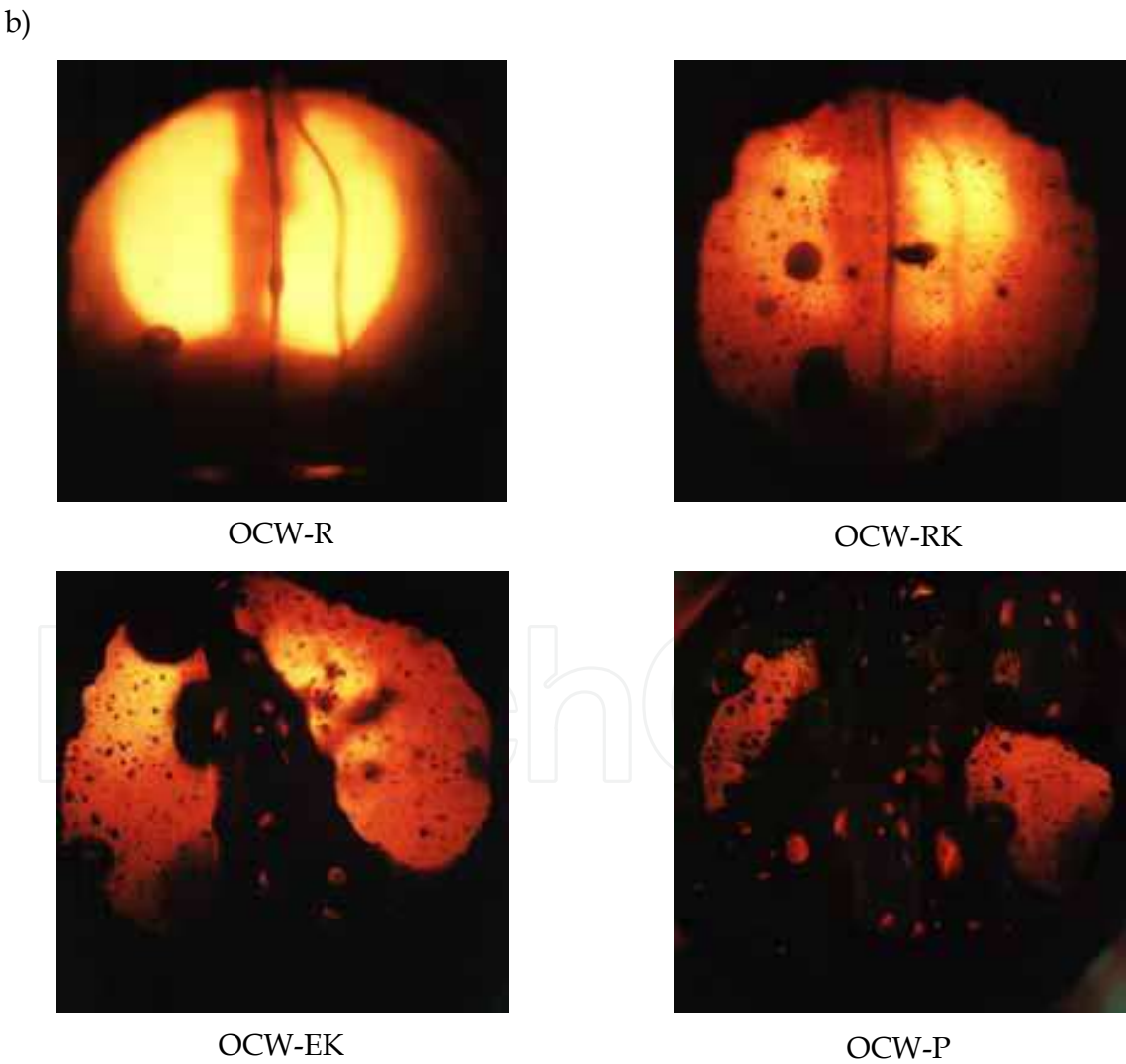
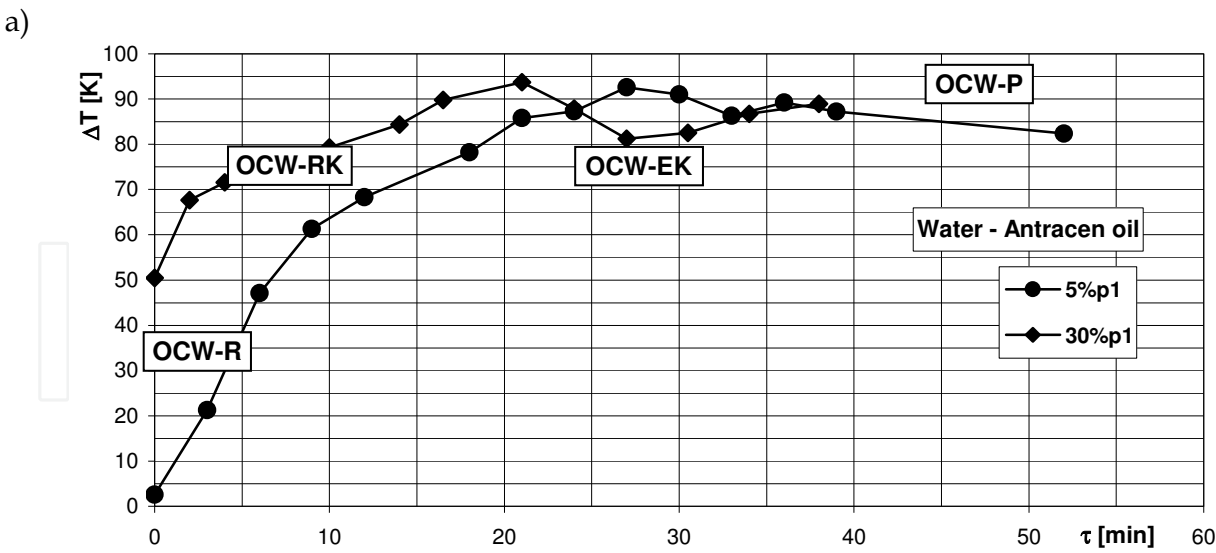


Fig. 14. View run-time of water – anthracene oil boiling mixtures: a) temperature difference; b) image structures ($\epsilon_{ol}=5/15\%$, $q=37\text{ kW/m}^2$)

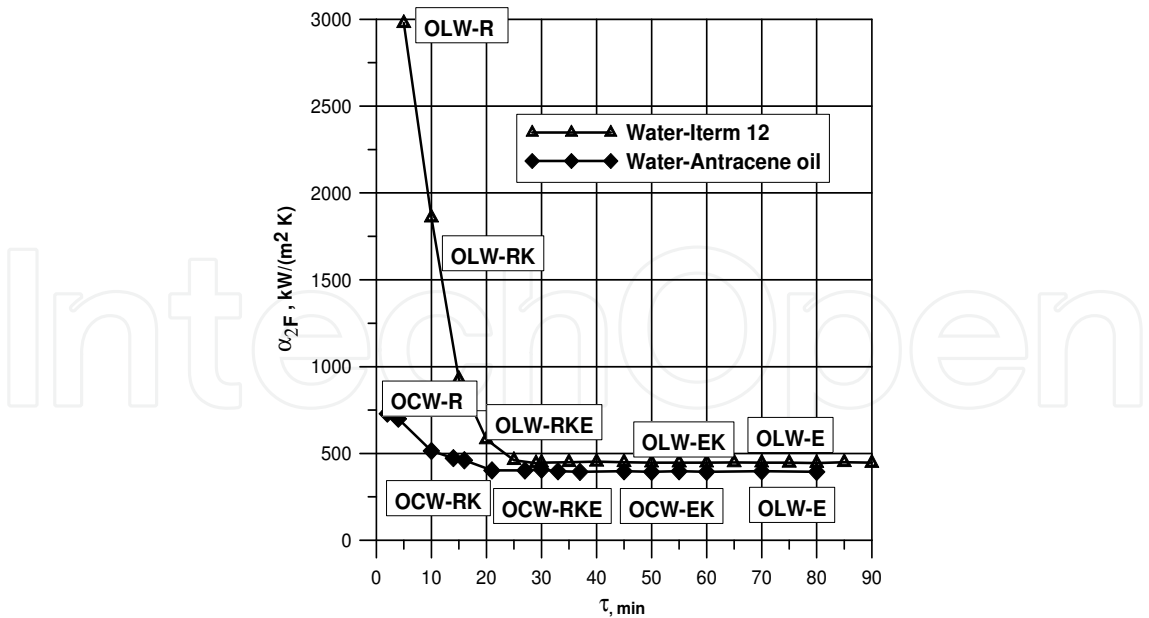


Fig. 15. Transient performance of boiling curves for water-oil mixtures: OLW - $q=26,7$ kW/m², $\varepsilon_{ol}=12\%$; OCW - $q=37$ kW/m², $\varepsilon_{ol}=15\%$);

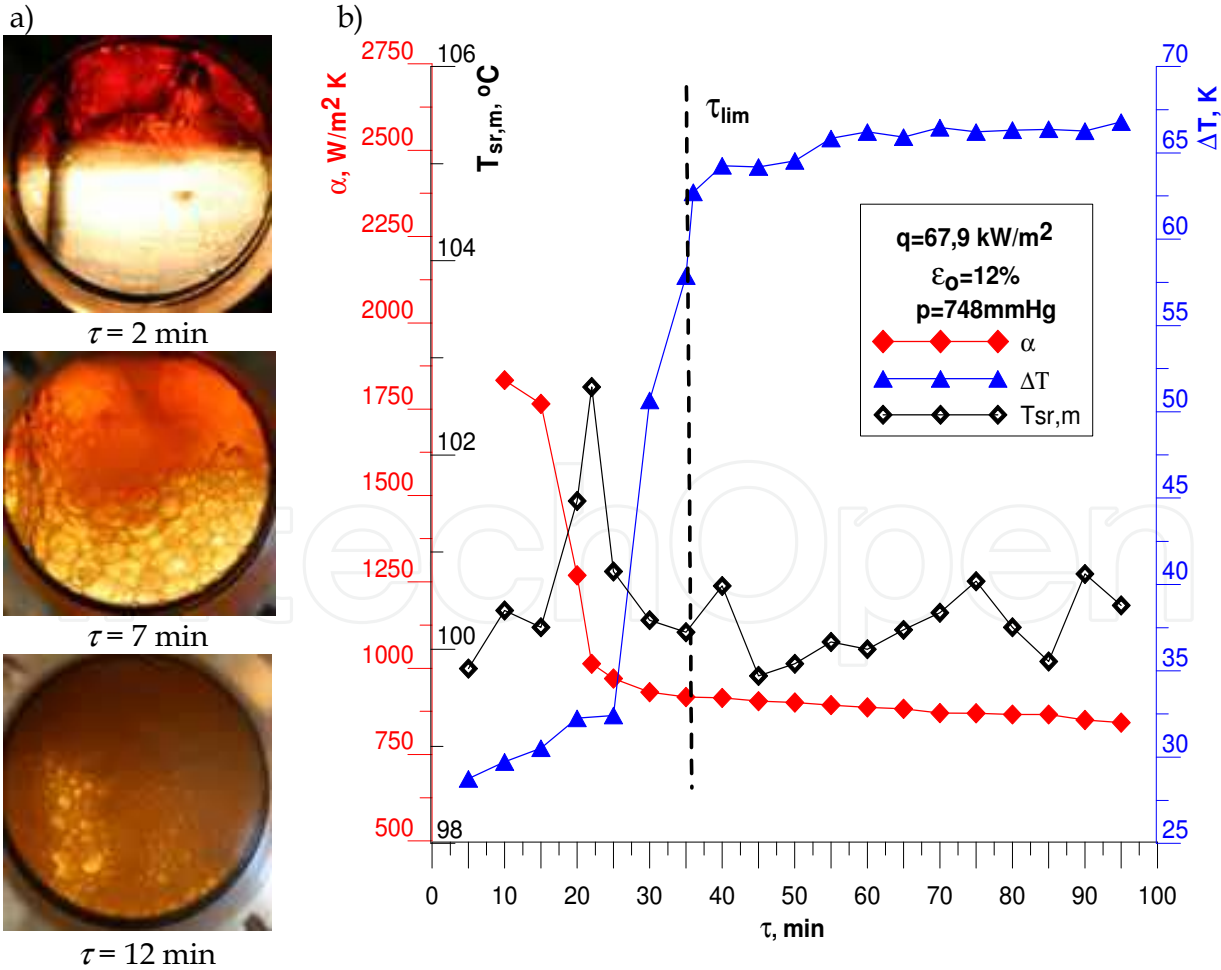


Fig. 16. Run of events (a) and boiling curves for water-oil item 12 mixture (b); $\varepsilon_{ol}=12\%$ vol.

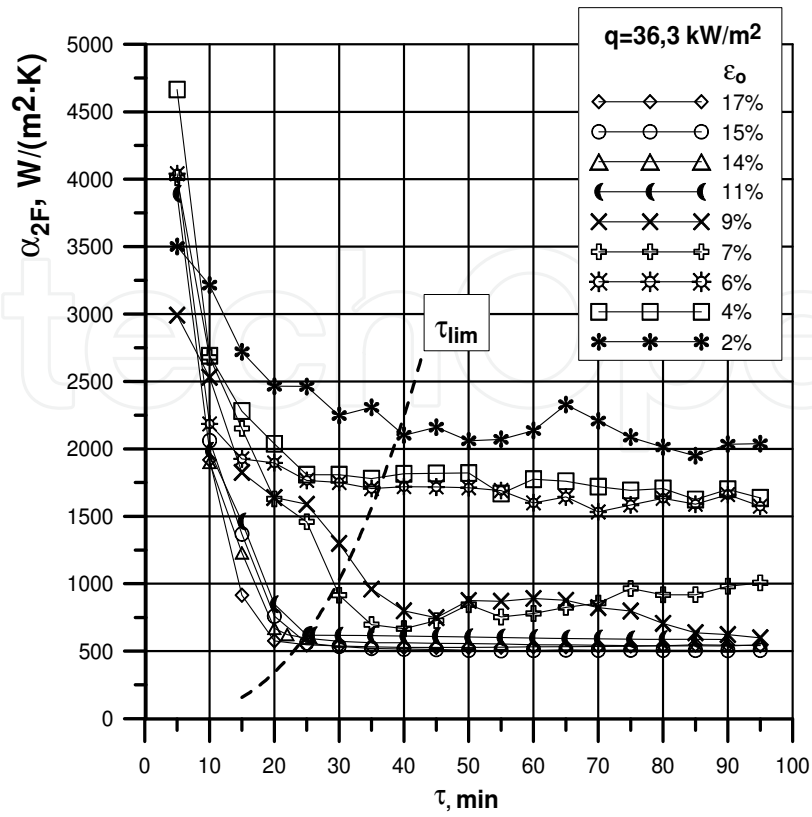


Fig. 17. Run-time of heat transfer coefficient; water-iterm oil 12 mixtures ($q=36,3 \text{ kW/m}^2$)

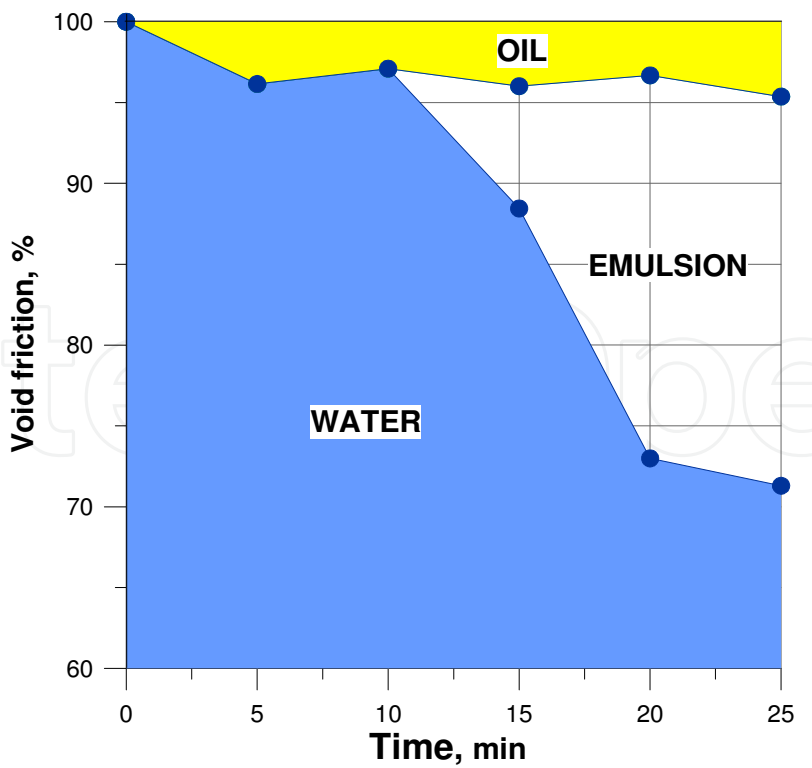


Fig. 18. Change of phases concentration near to the heating surface ($q=26,7 \text{ kW/m}^2$, $\epsilon_{ol}=15\%$)

It should be note - what shown in Fig. 18 - that the increase of oil concentration on the heating surface occurs successively with the boiling time, and depends both on the heat flux (q) and the initial oil content in water-oil mixture (ϵ_{ol}). Unsteady flow of heat transfer is revealed by a continued increase the temperature of heating wall, which is the higher than greater concentration of oil at the surface. Resulting from these conditions the temperature difference (from the definition of temperature between the plate and the boiling point of water) set at a level (30-100) K. Higher values were obtained for increasing values of heat flux (q) and increasing value of oil concentration (ϵ_{ol}).

It was found that the value of the temperature difference ΔT between the temperature of the heating wall surface and the saturation temperature of water determines the formation of specific structures point, that is: for $\Delta T < 15K$ - heterogeneous structures are formed (OLW-R, OLW-RK), in the range $15 < \Delta T < 40K$ - structures with dynamic emulsion (OLW-RKE, OLW-EK), and at $\Delta T \geq 40K$ - structures with permanent emulsion OLW-E type. These observations suggest that in real systems it is possible to control the boiling process of water-oil mixtures, by setting the required temperature of the heating medium.

The existence of these transition regions (ΔT overshooting) find reflection in the heat transfer coefficient values as shown in Fig. 19. The reduction of heat transfer coefficient (α_{2F}) value together with increasing of oil fraction in mixture is connected probably with dominant influence of the free convection in oil phase on the heat transfer. It should be emphasised, that the similar reduction of α_{2F} value is observed during flow boiling process of the oiled homogeneous substances, such as ammonia or dewatered coal tars (Filipczak, 1993, 2009; Witczak, 1997).

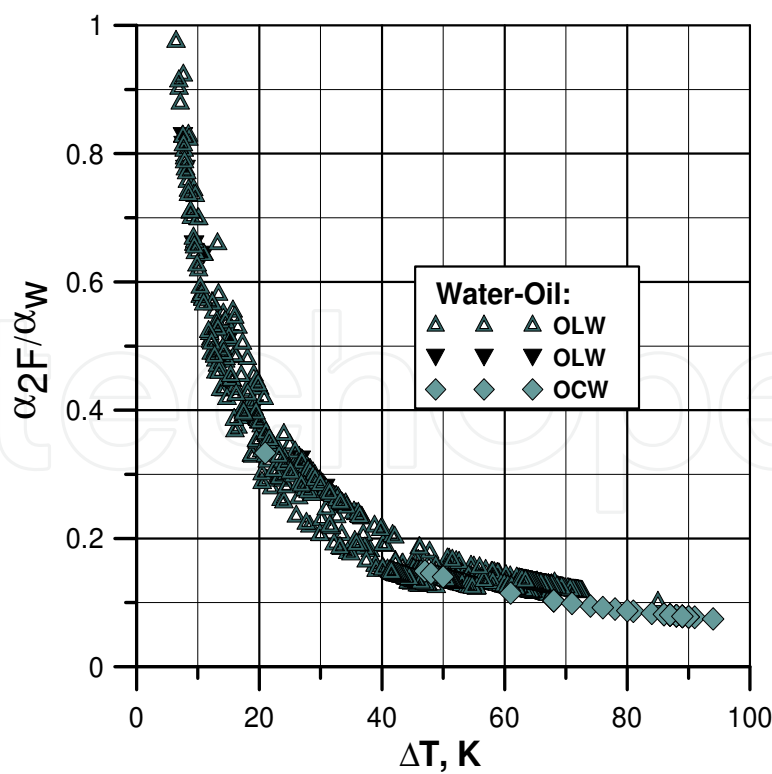


Fig. 19. Heat transfer damping effect during boiling of water-oil mixtures in relation to pure water – area of the research

The effect of damping, i.e. reduction - in relation to water boiling - of heat transfer rate during the water-oil mixture boiling has been shown in Fig. 19. As results from the line schedule (average relations), even a small fraction of oil on heating plate causes the strong lowering of the heat transfer coefficient. At the same time the higher is value of heat flux density (also, where the mixture can be in the form of emulsion or dynamic foam), the more significant is α_{2F} value reduction. It will be emphasised that the higher is oil volume fraction in mixture, the higher is effect of heat transfer coefficient reduction. For the highest fraction of oil investigated the heat transfer coefficients are stabilising on an approximately constant level, near the values determined by the free convection in oils component (Fig. 9).

Appointed on the investigation results the border condition describing limited time which indicates change of heat transfer conditions in the area of phase inversion was found experimentally as

$$\tau_{lim} = \frac{2.18 \cdot 10^8}{q^{1,14} \varepsilon_{ol}^{0,11} \left(\frac{D}{H} \right)^{0,27}} \quad (1)$$

where relation D/H refers to diameter of heating surface (D) and high of bulk water-oil mixture (H) respectively.

Basing on assumption that heating surface area is entirely covered by water-oil mixture, advantageously by emulsion, and it staying proportionally to oil and water concentration near thermal boundary layer (ε_{ol}^* , $(1 - \varepsilon_{ol}^*)$, respectively), the total temperature gradient is provided by components ratio, i.e.:

$$\Delta T = \varepsilon_{ol}^* \Delta T_{ol} + (1 - \varepsilon_{ol}^*) \Delta T_w \quad (2)$$

Take into consideration that boiling process proceeds on uniform heat flux (q) by simple calculation we obtained the relation for total heat resistance as

$$\frac{1}{\alpha_{2F}} = \frac{\varepsilon_{ol}^*}{\alpha_{ol}} + \frac{1 - \varepsilon_{ol}^*}{\alpha_w} \quad (3)$$

Hence,

$$\frac{\alpha_{2F}}{\alpha_w} = \frac{1}{1 + \varepsilon_{ol}^* \left(\frac{\alpha_w}{\alpha_{ol}} - 1 \right)} \quad (4)$$

Parameter ε_{ol}^* indicates a set of oil concentration near heating surface and its value may be from domain $0 \leq \varepsilon_{ol}^* \leq 1$. When oil phase is absent from heating surface, what take place for stratified or more separable water-oil structures (see Fig. 2a, Fig. 3a), than $\varepsilon_{ol}^* = 0$ and heat transfer coefficient converges to $\alpha_{2F} = \alpha_w$. On the other hand, for $\varepsilon_{ol}^* = 1$ oil liquid occupies all space of heating surface area (Fig. 3d, Fig. 4d, Fig. 5), and thus $\alpha_{2F} = \alpha_{ol}$.

It needs underlined that heat transfer coefficient a_W put in equation (4) for water boiling should be calculate from

$$\alpha_w = 3,15q^{0,7} \quad (5)$$

as the best results of own experiments. Likewise, to aim calculation of heat transfer coefficient for natural convection and nucleate or transition boiling of oil phase on horizontal plates the new equation, similar to Michiejew view one, is proposed, as

$$Nu_{ol} = 1,9Ra_{ol}^{0,25} \left(\frac{Pr_{ol}}{Pr_{th}} \right)^{0,25} \quad (6)$$

Equation (6) was verified for Rayleigh number of oil (Ra_{ol}) from $1,06 \cdot 10^8$ to $6,8 \cdot 10^{10}$.

The empirical correlations useful for calculation of oil concentration at heating surface are as follows:

- for non-steady conditions, i.e. for range restricted to separated or strongly heterogeneous structures of water-oil mixtures

$$\varepsilon_{ol}^* = 0,0368\varepsilon_{ol} \left(\frac{q}{1000} \right)^{1,47} \left(\frac{\tau}{3600} \right)^{1,33} \left(\frac{D}{H} \right)^{0,333} \quad (7)$$

- for steady run of process that is quasi-homogenous structures or fully development intermixed water-oil system and emulsions

$$\varepsilon_{ol}^* = \left[\varepsilon_{ol}^2 + 0,11 \frac{(1-\varepsilon_{ol})}{\varepsilon_{ol}} \left(\frac{q}{1000} \right)^{0,21} \right]^{-1} \quad (8)$$

what is rightly over the limited time given by equation (1).

At occurrence of intermediate or transitions structures the calculate programme according to heat transfer coefficient by equation (4) needs the less value of oil concentration ε_{ol}^* obtained from equations (7) and (8).

The computation results supported by statistic analysis show that in range of done experiments the proposed method describes heat transfer coefficient with mean error no more than 25% and may be used to prediction of heat transfer conditions upon pool boiling of water-oil mixtures different kinds. The results of this analysis indicate as well that irrespective of components concentration in water-oil systems with the help of equation (1) may evaluate the time for advantageous run of water-oil mixture boiling process.

5. Conclusions

Based on results of experimental investigations the following most significant conclusions can be formulated:

1. The process of boiling water-oil mixtures exhibits a number of peculiarities. They result from the diversified structure of liquid-liquid system and manifest themselves in a sudden variability of the process parameters.
2. The nature of the boiling process depends on the kind and composition of water-oil mixtures. Watering of mixtures favours the process of nucleation and contributes to quicker shattering of oil phase in the bulk. With a high of oil content, the heat transfer coefficient is dominated by free convection in the oil phase what leads to sudden decrease of heat transfer efficiency.
3. Depend on composition of water-oil mixture the heat transfer conditions are mirror of the steady as well as non-steady criteria and conditions. The first take place for separated form of water-oil mixture and second ones are beginning at quasi-homogeneous structures of this mixture (e.g. emulsions). The oil content has a very considerable effect on boiling curve then.
4. As the result of investigations, the phenomena connection occurring during pool boiling of two-phase water-oil mixture and the influence of time condition on process run of events and its parameters the relations for the value of heat transfer coefficient was worked out.
5. Description and structures systematics of boiling water/oil/water systems can greatly facilitate for further work on this process.

Symbols

D	– diameter of heating surface, m
H	– high of oil-water mixture (total level)
Nu	– Nusselt number
Pr	– Prandtl number
q	– heat flux, W/m^2
T	– temperature, K
ε_{ol}	– void fraction of oil
ε_{ol}^*	– concentration of oil
α	– heat transfer coefficient, $W/(m^2 \cdot K)$
t	– time, s

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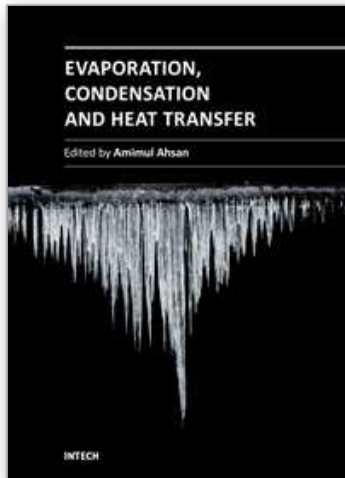
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