Heat transfer under powerful heat release in supercritical-pressure liquids

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Abstract. The aim of the present article is to summarize the results of pulse experiments performed under conditions of powerful heat release in supercritical-pressure liquids. The parameters of the discussion are the values of the speed of crossing the vicinity of the critical temperature and pressure reduced to the critical one. These results obtained for “heater-probe / supercritical fluid” system and based solely on comparison of the primary data indicate a threshold decrease in the intensity of heat transfer near the critical temperature. The purpose of this summary is to suggest a relatively simple approach to reconciling the pulse heating results with the peaks of thermophysical properties derived from experiments in stationary conditions. It is assumed that the action of gradient in temperature and the presence of heat-transfer surface can serve as factors that suppress large-scale fluctuations in pulse heated system, leading to a “smoothing” the critical enhancement of the properties.

1. Introduction
Within the nuclear electricity production industry, the principal task is to eliminate emergency events. This fully applies to the development of power units based on supercritical heat carriers. Indeed, the prospects of supercritical fluids (SCFs) are justified, since a boiling crisis is impossible at supercritical pressures (SCP), which allows increasing the thermal efficiency by raising the temperature of the supercritical heat carrier. Moreover, the properties of fluids show an extreme behavior directly in the near-supercritical region [1, 2].

On the other hand, heat transfer deterioration (HTD) mode, which manifests itself starting at a certain heat flux density value, makes these prospects less than obvious. HTD mode, which was discovered more than 50 years ago [3], continues to be an obstacle to the use of SCFs in the processes, in which high-power local heat release appears to be possible. In order to perform this study, it is of fundamental importance to elucidate the peculiarities of heat conduction in a viscous sublayer at significant heat flux densities. Such information is expected to clarify the boundaries of the effects of the peaks of thermophysical properties on the heat conduction of SCF for the growing range of variation of the heat flux density. In recent years, there has been a paradoxical situation, when the notion of the peaks of thermophysical properties, known from stationary measurements and listed in the Standard Tables [4], peacefully coexists with the results of pulse experiments performed under conditions of high-power heat release. These results obtained for “probe/SCF” system in the framework of the high-speed technique [5] and based solely on comparison of the primary data indicate a threshold decrease in the intensity of heat transfer near the critical temperature [6–10].
An explanation of this situation becomes a fundamental scientific task. The aim of the present article is to summarize the results of pulse experiments performed in conditions of high-power heat release. The parameters of the discussion are the values of the speed of crossing the vicinity of the critical temperature and pressure reduced to the critical one. The purpose of this summary is to suggest a relatively simple approach to reconciling the pulse heating results with the peaks of thermophysical properties derived from experiments in stationary conditions.

2. Background
Due to the anomalous behaviour of thermophysical and kinetic properties of substances in the near-critical region, SCFs have attracted considerable attention both in terms of their inherent interest to science and their various technological applications in industrial processes, see [11, 12] and bibliography therein. From an experimental point of view, supercriticality leads to elevated requirements both in terms of the procedure for carrying out the primary measurements, as well as the correspondence of the experimental conditions to the ideal model for converting the primary data into values representing the substance's thermal properties. With respect to the heat transfer in supercritical fluids, the following points have been formulated and generally accepted [1, 2, 4]:

• the heat capacity values have distinct maxima along the supercritical isobars;
• the thermal conductivity values show similar behavior;
• the heat transfer coefficient values have a maximum in the near-supercritical region.

It is accepted that the existence of peaks owes its nature to large-scale fluctuations, as well as increased convective mobility in the third case. Indeed, this set of considerations forms the basis for “heat transfer enhancement” mentioned in the scientific literature. However, the results of heat flux density measurements have revealed the following problem: the peak of heat transfer coefficient reduces as the heat flux density increases, see [10] and bibliography therein. As a result, the following contradiction is observed. The removal of small heat fluxes, corresponding to small temperature differences at which the peak “works”, is not subject to an engineering solution. Rather, the problem for engineers lies in the safe removal of high-power heat fluxes. Thus, the justification for the use of supercritical fluids in prospective nuclear power programmes is not definitive and should be the topic of further study.

3. Experimental
In order to study non-stationary heat transfer in SCFs, the method of controlled pulse heating of a wire probe (resistance thermometer) [5] was applied. Here only the essence of the method is pointed out. The method relies on the use of device for the selection of the heating function combined with device for recording the corresponding response function over the heating range. The parameters of the heating power $P(t)$ are defined by the conditions of the problem. In practical terms, the most significant specific case is the constant heating power mode. Correspondingly, the response function is the resulting heating profile $T(t)$ for the given $P(t)$, as shown in figure 1. The higher the response curve $T(t)$ for the given power value, the lower the intensity of heat transfer by a given sample. These data are sufficient for determining the heat flux density through the probe surface $q(t)$, which is the most important observable in transient heat-transfer experiments with large changes in the substance temperature

$$ q = \frac{(P - P_0)}{\pi \cdot d \cdot l} $$

(1)

and the thermal resistance of a substance at a given heating mode

$$ R_i(t) = \frac{\Delta T(t)}{q(t)} $$

(2)

These values are calculated directly from the primary data. Here $d$ and $l$ are the probe diameter and its length, respectively; $\Delta T(t)$ is the temperature rise; $P_0(t)$ is the fraction of the applied power consumed for heating of the probe itself.
Figure 1. Heating curves of the probe and their derivatives with respect to time $\partial T/\partial t$ (insert) for isopropanol, $p_c \approx 4.9$ MPa, $T_c \approx 236^\circ C$, under constant power heating mode. The values of $p/p_c$ from 1.02 to 6.0 serve as a parameter. The isobars in the $(1\div3)p_c$ interval are given by dashed lines. The sensitivity to pressure changes at $p < 3p_c$ was approximately 0.05 MPa, at $p > 3p_c$ – 0.1 MPa.

The pulse experiments consisted in the rapid isobaric transfer of compressed liquid to the region of supercritical temperatures over a wide range of supercritical pressures from 1 to $(2\div7)p_c$. The characteristic heating time was measured in milliseconds, corresponding to the thickness of the heated layer in microns. The results provide information on heat transfer under the predominance of the heat conduction mechanism. They indicate a threshold decrease in the intensity of heat transfer in the vicinity of the critical temperature at $(1\div3)p_c$ [6–10], see figure 1. Let us note two points inherent in this approach. The first relates to the obtained spatial scale being a good approximation precisely for the viscous sublayer. The second rests in the fact that the temporal scale allows practically excluding the influence of gravity and convective instability effects.

Under such conditions, a change in the probe temperature $T(t)$, which is recorded in the experiment, can directly correlate with changes in the intensity of heat transfer of the substance under a given heating power. By changing the external parameter (in our case, pressure), it is possible to observe changes in the heat transfer patterns associated with changes in the parameter at sub- and supercritical temperatures. Here it is of central significance that the heating power history $P(t)$ is repeated with sufficient accuracy over the entire series. This serves as a basis for the transition to the relative mode of thermal response measurements.

Experiments on unsteady heat transfer with entering the region of supercritical parameters were carried out for liquids of various chemical natures. The liquids stated were alcohols, ketones, esters, and water. The data obtained have revealed a high degree of main result universality, namely, a significant increase in thermal resistance as the pressure approaches (from the above) to the critical one. In figure 2, this result is presented in a qualitative form.
Figure 2. Thermal resistance $R_{ij}$ reduced to thermal resistance at the highest pressure $R_{0i}$, depending on the reduced pressure $p/p_c$ for various substances: 1 – isopropanol, 2 – malonic diethyl ether, 3 – acetone, 4 – water. Data for water in the range of $p/p_c$ values from 3 to 5 were obtained by calculation [8] using data [4].

Not only does this observation contradict the heat transfer pattern typical of the stationary case, it is also at odds with the predictions of the theory of critical phenomena. Therefore, the following approach was chosen as the first step in reconciling the results of pulsed and stationary experiments. The decision was taken to gradually increase the pulse duration in our measurements until the appearance of signs of convection, thereby reducing the gap between the experimental conditions under discussion. In our heat-pulse experiments, the passage time of the temperature region corresponding to the isobaric heat capacity peak was 0.1 ms by the order of magnitude. Taking the short heating duration into account, it was important to perform a special experiment; here, the systematic extension of the passage time was of particular interest. In the presented experiments, we managed to decrease the rate of passing the temperature range of interest by an order of magnitude with respect to previous case [6–10]. The increase in time period gave a similar result, but this was already against the background of signs of free-convective heat transfer following critical transition, see, for example, figure 3. No effect of the thermal property peaks on heat transfer was detected.

4. Discussion

It was previously found that the effect of the threshold decrease in the intensity of heat transfer is enhanced when approaching the critical pressure, while its resolution ceases at $p/p_c \rightarrow 3p_c$. Such an extended effect along the pressure axis contrasts sharply with the known results of experiments on measuring the thermodynamic and transport properties in the vicinity of the critical point and could not be easily predicted in advance. At the same time, this finding is not unique: a similar result was reported by D.Yu. Ivanov on the example of the thermal conductivity of ammonia [13]. The nonmonotonicity of the thermal conductivity (with respect to density) curve in the vicinity of the critical isochore could be clearly traced very far from the critical point ($T > T_c$) – in fact, at a distance of around a hundred degrees.
Figure 3. Consecutive pulses of water heating along the isobar of 23 MPa (curves 1, 2, 3) at selected power mode (insert). The arrows show the timing of supercritical transition (I) accompanied by the effect of the threshold decrease in the intensity of heat transfer and free-convective transfer onset (II).

A very informative component of the discussion consists in the fact that the response signals in pulse experiments on individual liquids do not contain any features in the range of $p > 3p_c$. The pattern of heat transfer in this region resembles that for weakly compressible subcritical fluids. In figure 1, this statement is illustrated by the example of isopropanol. In the $p > 3p_c$ region the response signals reflect the natural tendency to increase the substance thermal effusivity with pressure. For clarity, the response signals for isobars in the $(1\div3)p_c$ interval are given by dashed lines. The experimental results on the heat conduction of SCF, which were found under conditions established by us, appear to be paradoxical when taking the above mentioned “generally accepted points” into account. Indeed, as soon as non-stationary conditions were set against the background of significant gradients of physical variables, which is equivalent to small characteristic times and sizes, the SCF phenomenon changed its “familiar face”.

Attempts to understand the physical nature of the observed phenomenon naturally lead to characteristic features concerning the structure of SCF. Apparently, the structuring factor becomes the main reason for the threshold decrease in the intensity of heat transfer, which was clearly manifested in our experiments. It is accepted that in the near-supercritical region, the substance loses its structural homogeneity [14]. Let us mention two circumstances directly related to the problem. Firstly, gas-like domains remove the heat less effectively. It is to be supposed, as a part of the ongoing discussion, that this circumstance is a fundamental factor in the emergence of the HTD mode [15]. Secondly, nearly any violation of the homogeneity of the system, according to experimental experience (see, for example, [16]), serves as an additional barrier to heat transfer. On the other hand, these factors can not affect the dominant role of the peaks of thermodynamic and transport properties. It is reasonable to assume that a substance entering the region of near-critical values of $T$ and $p$, which are set in the pulse experiments, does not pass into a critical state in the usual sense of the term.
Under conditions of large values of the heat flux density and temperature gradient, as well as a short response time of the heat-transfer surface, the long-range critical correlations are not formed. Accordingly, anomalies of the properties well-known from stationary measurements do not manifest themselves. The possibility of reaching a state with a small amplitude of fluctuations and, as a consequence, with nonsingular thermodynamic functions at sufficiently rapid penetration into the near-critical region was predicted by Zel’dovich [17]. In the case of the experiments discussed, two more factors, namely the temperature gradient and the presence of a solid surface, that impede the establishment of complete equilibrium of correlations [18] are added to the short-term factor.

Conclusions

The application of a short-term technique for studying supercritical heat transfer has led to unexpected results capable of influencing the direction of future research on this subject. The short thermal relaxation time of the probe allowed identifying heat transfer patterns at small characteristic times and sizes, as well as at high heat flux densities. Within this approach, the impact of the two main factors complicating the study of heat conduction in SCFs has been practically eliminated. As a result, the effect of threshold decrease in the heat transfer intensity and its sphere of influence with respect to the change in reduced pressure values was found. Thus, the experimental conditions were found, accompanied by suppression of large-scale fluctuations in thermodynamically non-equilibrium system and, consequently, smoothing of the critical enhancement of the properties. It is surprising that this effect, which is rather significant for understanding the essence of supercritical heat transfer, has not received a clear response in the seven years since the first publications on this topic [6–8].

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