Thermal Waves, Thermal Diffusivity and Possibility of Relaxation Time of Materials Evaluation

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Abstract

In the paper the evidences of existing thermal waves during quenching steel probes in liquid media are provided. It is supported by theoretical consideration and accurate experiments. Practically, thermal waves can be depicted by thermocouples if quenching is performed in electrolytes. In this case, the double electrical layer acts like an amplifier for thermal wave distribution. The double electrical layer eliminates full film boiling process and is responsible for self – regulated thermal process establishment. The speed of thermal wave distribution and duration of self – regulated thermal process are used for thermal diffusivity and relaxation time of materials evaluation. Obtained by such a way results are used for film boiling prediction, initial heat flux density calculation and quenching recipes development. The obtained in the paper results can be used by engineers and scientists dealing with investigation of physical properties of materials.

Keywords - *Thermal waves, double electrical layer, speed of wave, thermal diffusivity, relaxation time, initial heat flux density, recipes, practical use.*

I. INTRODUCTION

It is widely distributed opinion that during quenching of heated to high temperature steel parts in liquid media always three phase of cooling take place: film boiling, nucleate boiling and convection. Many well known scientists and investigators are bonded by this opinion and considered such situation as classical fact that should be never changed both at present and in the future. It is based on many experimental facts and heat conductivity conventional theory which says that during immersion of heated to high temperature steel parts into liquid initial heat flux density tends to infinity. If so, initial heat flux density always prevails the first critical value q_{cr1} and it means that film boiling is always present in vaporizable liquid media. The first scientist who made a difference was French [1]. He prepared spherical samples of different diameters made of AISI 4140 steel which were accurately instrumented by thin surface thermocouples shown in Fig. 1.

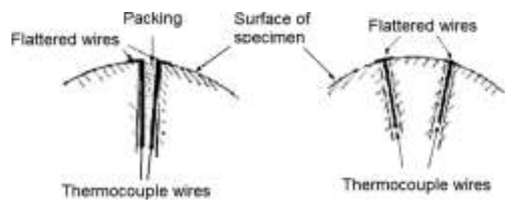


Fig. 1: Depiction of how thermocouples were arranged and accurately flattened to the wall of spheres and polished by French [1]

He quenched spherical probes from 875 °C in 5 % water solution of NaOH at 20 °C agitated with 0.914 m/s (French, 1930) [1]. Some results of his numerous experiments are presented in Table 1.

TABLE I
Time required for the surface of steel spheres of different sizes to cool to different temperatures when quenched from 875 °C in 5 % water solution of NaOH at 20 °C agitated with 0.914 m/s (French, 1930)

Size, Inches, (mm)	**Time, sec**							
	700°C	600°C	500°C	400°C	300°C	250°C	200°C	150°C
0.5” (12.7)	0.028	0.042	0.058	0.071	0.11	0.15	0.26	0.60
4.75” (120.6)	0.043	0.066	0.09	0.12	0.17	0.21	0.29	0.95
7.15” (181.6)	0.040	0.070	0.100	0.140	0.240	0.310	0.42	1.15
11.25” (285.8)	0.043	0.120	0.190	0.330	0.570	0.960	1.26	2.18

As one can see from Table 1, there is no film boiling at all during quenching spherical probes in 5 % water solution of NaOH at 20 °C.

Thermal scientists didn't pay serious attention to these accurate experiments of French because the data somewhat contradicted existing theory. In this paper experimental data of French were used to solve inverse problem (IP) to see initial heat flux densities during immersion of steel probes into liquid quenchant. Currently, technique of solving IP is highly developed [2, 3]. Author used IQLab program [4] which is based on parabolic heat conductivity equation with a third kind of boundary condition. Results of computer calculations are presented in Fig. 2 and Fig. 3 a), b).

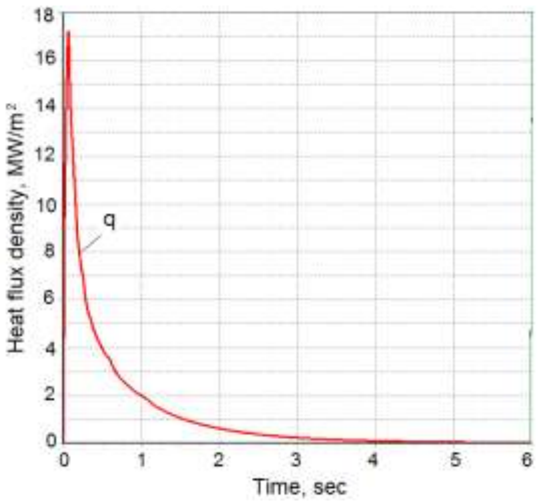
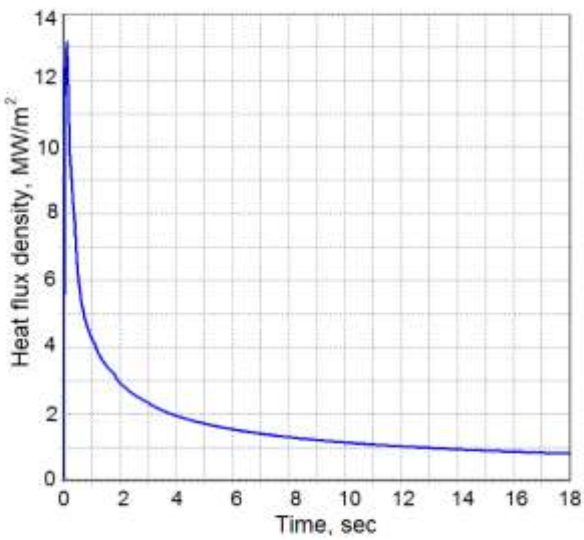
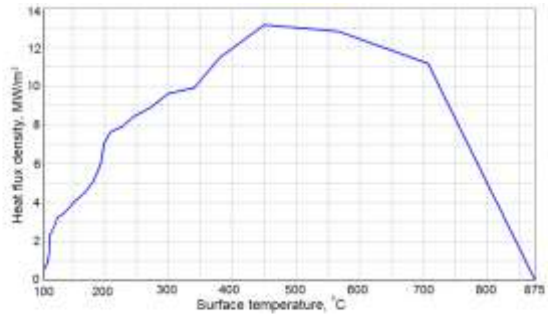


Fig. 2: Heat flux density versus time during quenching spherical steel probe 12.7 mm in diameter in 5 % water solution of NaOH at 20°C agitated with 0.914 m/s.



a)



b)

Fig. 3: Heat flux density during quenching spherical steel probe 181.6 mm in diameter from 875°C in 5 % water solution of NaOH at 20 °C agitated with 0.914 m/s: a) is heat flux versus time; b) is heat flux versus surface temperature.

As seen from calculated data, initial heat flux density is rather high and it looks like it accedes the first critical value q_{cr1}.

II. THERMAL WAVES TAKING PLACE DURING QUENCHING

To solve the problem connected with the theoretical non realistic infinity of the initial heat flux density and infinity of heat distribution, Vernotton and Lykov proposed to use modified heat conductivity law of Fourier (1) instead of already existing conventional one [5, 6]:

$$q = -\lambda \frac{\partial T}{\partial r} - \tau_r \frac{\partial T}{\partial \tau} \qquad (1)$$

Here λ is thermal conductivity of material in W/mK; τ_r is relaxation time in s. If relaxation time τ_r is extremely small, the value of τ_r tends to zero and we have conventional heat conductivity law of Fourier (2)

$$q = -\lambda \frac{\partial T}{\partial r} \qquad (2)$$

As known [6], modified heat conduction law of Fourier (1) generates hyperbolic heat conductivity equation (3):

$$\frac{\partial T}{\partial \tau} + \tau_r \frac{\partial^2 T}{\partial \tau^2} = a\nabla^2 T \qquad (3)$$

Multiplying equation (3) by $c\rho V$, we get

$$c\rho V \frac{\partial T}{\partial \tau} + \frac{\lambda V}{w_r^{\,2}} \frac{\partial^2 T}{\partial \tau^2} = V\lambda\nabla^2 T \qquad (4)$$

Since the mass ρV of free electrons in metal is extremely small, one can state

$$c\rho V\frac{\partial T}{\partial \tau} \to 0 \qquad (5)$$

Assume that $c\rho V\frac{\partial T}{\partial \tau} = 0$, and then we receive the well – known equation for thermal wave distribution (6)

$$\frac{\partial^2 T}{\partial \tau^2} = w_r^{\,2}\nabla^2 T \qquad (6)$$

According to author [6], w_r is speed of heat distribution in m/s and it can be calculated by Eq. (7) if thermal diffusivity a of material is known:

$$w_r = \sqrt{\frac{a}{\tau_r}} \qquad (7)$$

From the above consideration follows the wave behavior of free electrons during metal quenching in liquid media where during immersion into liquid a huge temperature gradient is formed and double electrical layer is established [7].

To be more specific, hyperbolic heat conductivity equation (3) should be completed by the boundary and initial conditions. In 1973 authors [8] proposed the boundary condition for transient nucleate boiling process using Tolubinsky's dimensionless equation (8) [9, 10]:

$$Nu = 75K^{0.7}\,\mathrm{Pr}^{-0..2} \qquad (8)$$

After reducing Eq. (8) to more simple form (9),

$$\alpha_{nb} = Cq^{0.7} \qquad (9)$$

authors [8] received the boundary condition for transient nucleate boiling process in the form (10):

$$\left[\frac{\partial T}{\partial r} + \frac{\beta^m}{\lambda}\left(T - T_s\right)^m\right]_S = 0 \qquad (10)$$

Initial condition for quench process is

$$T(r,0) = T_o \qquad (11)$$

III. FILM BOILING ABSENCE EXPLANATION FROM THE POINT OF VIEW OF PHYSICS

The experimental results from French's studies are summarized in Table 1 which shows that during quenching spherical steel samples in a cold 5 % water alkaline solution film boiling is absent. Also, independently of size of sphere, the wall temperature decreases very quickly from 975°C to 150°C within one second which would seem impossible. However, using computational methodology, such as finite element analysis, it is possible to solve this heat transfer problem as an inverse problem (IP). Details and methods of solving the inverse problem are provided in Chapter 13 [11]. Some results of computations using the software IQLab [4] are shown in Figs. 2 and 3. Fig. 2 shows that the heat flux density is very high at the very beginning of the quenching process and varies smoothly as the process proceeds. Authors [11] showed that the maximum heat transfer coefficient (more than 200,000 W/m^2K) is achieved at the time of 2 seconds. Furthermore, the heat transfer coefficient decreases exponentially and reaches 30,000 W/m^2K within 20 seconds after immersion [11].

Initial process of quenching is rather complicated and currently is not investigated deeply and widely yet. As discussed, investigators consider film boiling, nucleate boiling and convection during quenching or nucleate boiling and convection if film boiling is absent [12]. For each stage of heat transfer the well - known boundary conditions are used [11, 12]. However, during immersion heated steel part into cold liquid, first natural convection takes place since vapor bubbles are not formed yet. To start any boiling process, cold liquid must be heated to saturation temperature and then boundary layer should be overheated that forms boiling centers. Possible two ways of heat transfer processes, during immersion heated to high temperature steel parts into cold liquid, are shown in Fig. 4 [12].

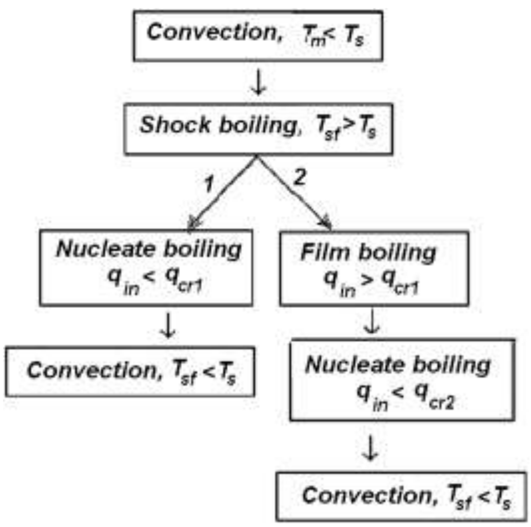


Fig. 4: The scheme of cooling process during hardening steel in cold liquid media [12].

For the first time shock boiling was discovered in 1997 by authors [13].

Thus, the full film boiling process is absent during quenching steel parts in cold liquid media when initial heat flux density q_{in} is below its critical value q_{cr1}. It is realistic because cold liquid must be heated first to saturation temperature and then went through the shock boiling process (see Fig.4 and Fig. 5) [13].

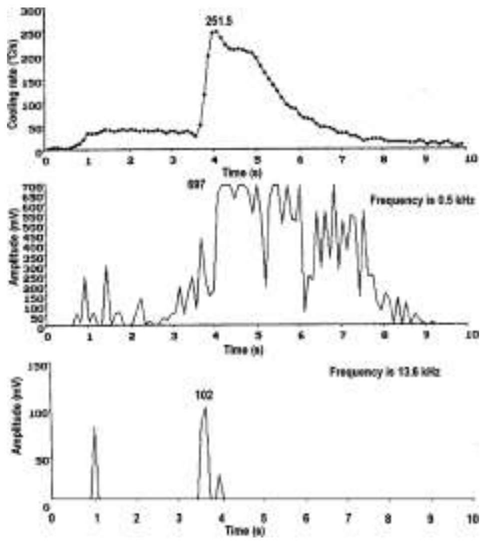


Fig. 5: Temperature–time, broadband, and narrowband quenching data [13].

As seen from Fig. 5, it is evident that at a frequency of 0.5 kHz there are appreciable sound effects from the oscillation of a vapor film during full film boiling. At a frequency of 13.6 kHz, only two sound spikes were observed, which were connected with the formation and growth of nucleating centers during shock boiling. Initially, vapor bubbles were very small, and the oscillation frequency was much higher than during developed nucleate boiling process. The first spike is due to the formation of nucleating centers that merged and created a vapor film. The energy connected with the oscillation of nucleating centers has passed to the oscillatory energy of a vapor film. The second spike is connected with the destruction of a vapor film and repeated formation of nucleating centers, which then have grown and begun to oscillate with less frequency. For conventional boiling process is true a ratio [9, 10]

$$\frac{q_{cr2}}{q_{cr1}} = 0.2 \qquad (12)$$

while for shock boiling the ratio (12) becomes as

$$\frac{q_{cr2}}{q_{cr1}} = 0.05 \qquad (13)$$

It means that during extremely fast cooling the first critical heat flux density increases almost four times as compared with conventional nucleate boiling that produces 2.5 – 2.8 mm bubbles oscillating with the frequency 56 – 67 Hz (see TABLE II).

TABLE II
Effect of heated surface material on bubble departure diameter and release frequency in the case of boiling water at normal pressure [9]].

Material	**d_o, mm**	**F, Hz**	**$W^{"} = d_o f$, s^{-1}**
Permanite	2.5	61	0.153
Brass	2.3	67	0.157
Copper	2.8	56	0.157

Thus, from the point of view of physics, during quenching steel parts in cold liquid media full film boiling can be absent.

IV. THERMAL WAVES RECORDED BY EXPERIMENTS

A. The Poker Effect

A long ago, metallurgists noticed that during immersion of heated poker into cold water results in a burning effect on another cold end of the poker. This effect is called a "poker" effect. The poker effect was explained for the first time by authors [14, 15]. However, nobody investigated properly poker effect to collect data concerning this unusual phenomenon. Currently, there are some theoretical and experimental data [14, 16] which were used in this paper to evaluate physical properties of materials.

B. Thermal waves affecting by a double electrical layer

To investigate the poker effect, a special probe made of carbon steel was prepared as shown in Fig. 6. A section of the thicker probe end having a length of 120mm was immersed into an electric furnace with the temperature of 500°C. Two different heating times were used: 30 and 120 minutes. Two thermocouples were located at the distance of 150 mm from the probe hot (left) end: thermocouple 1 at the core of the probe and thermocouple 2 on the surface of the probe (Fig. 6). Thermocouples 3 and 4 were located on the surface of the probe at the distance of 220 mm and 290 mm from the probe hot end [16].

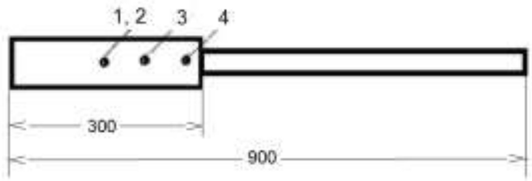


Fig 6: Probe made of carbon steel with locations of thermocouples in it [16]: 1 is a thermocouple located at the core at the distance of 150 mm from the left end of the probe, 2 is a thermocouple located on the surface of the probe at the same distance; 3 is a surface thermocouple located at the distance of 220 mm; 4 is a surface thermocouple located at the distance of 290 mm.

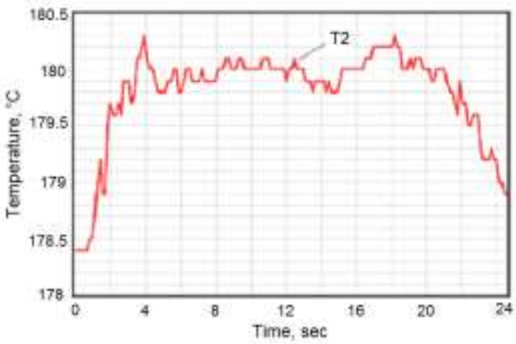


Fig. 7: Instant increase and oscillation of the surface temperature at a point 2 during cooling of the probe in 12% water solution of NaCl at 20°C which was immersed into solution immediately after heating (see Fig. 2)[16].

Different authors provide different relaxation time values. Lykov in his monograph [6] stated that relaxation time τ_r for aluminum is 10^{-11} s. It is so small value that he had a doubt on possibility to measure relaxation time in lab condition using contemporary equipment [6]. Prof. Buikis and his colleagues during analyzing and solving hyperbolic heat conductivity equation were dealing with the relaxation time of $\tau_r = 1.5s$ [17, 18]. Using experimental data provided in Fig. 7, it is possible to evaluate approximately relaxation time τ_r. The impulse time measured by measuring system with cylindrical probe (see Fig. 6) consists of two periods: probe transferring time and time needed for movement wave temperature signal within distance 0.15 m (see Fig. 6). Probe transferring time was approximately 0.5 – 0.6 seconds and temperature impulse was recorded by measuring system at a time 0.72 sec. If so, the speed of wave temperature movement is only $w = \frac{0.15m}{0.12\sec} = 1.25m/s$

It is rather small speed for movement temperature waves in metals. Keeping this fact in mind, let's evaluate for comparing relaxation time τ_r from equation $w_r = \sqrt{\frac{a}{\tau_r}}$.

Thermal diffusivity of probe material is $a = 14.8 \times 10^{-6} m^2/s$ (see Table IV). After substituting obtained data into above equation, the calculated relaxation time value is $\tau_r = 9.47 \times 10^{-7} s$. It is really small value even for temperature wave speed of 1.25 m/s. Exact experimental data can be received after complete automation of experimental measuring which can be functioned at the moment of touching of bottom of the probe with electrolyte that can immediately turn on measuring system. Also, distance for wave temperature signal movement should be extended up to 1 m. Along with improvement the experimental measurements, further investigations in the field of solving hyperbolic heat conductivity equation with the boundary condition taking into account existence of double electrical layer [7, 11] should be continued. Author of the paper believes that it can be done in the nearest future. At present, the problem on average thermal diffusivity of metals and iron evaluation is already solved using equation (14). Ukrainian Intensive Technologies Ltd Co., cooperating with Institute of Thermophysics of NANU, evaluated thermal diffusivities of Ductile Irons which are currently successfully used by Akron steel heat treating company in the USA. .

V. SELF – REGULATING THERMAL PROCESS

Duration of transient nucleate boiling is proportional to square of thickness of steel part, inversely proportional to thermal diffusivity of steel, depends on configuration of steel part quenched and cooling characteristics of quenchant if austenitizing and bath temperatures are fixed (see Eq. (14) and Fig. 6):

$$\tau_{nb} = \overline{\Omega} k_F \frac{D^2}{a} \qquad (14)$$

Fig. 8 provides the value Ω versus Biot number Bi depending on size of steel part and heat transfer coefficient during convection [19, 20].

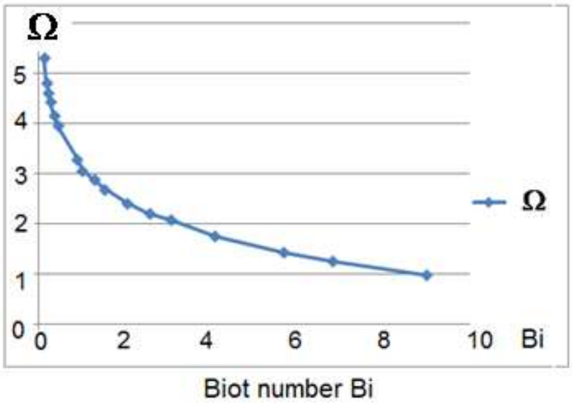


Fig. 8: Value Ω versus Biot number Bi depending on size of steel part and heat transfer coefficient during convection [20].

Form coefficients k_F for different shapes of steel parts and thermal diffusivity of selected metals are available from Table III and Table IV [21].

TABLE III

Form coefficients k_F for different shapes of steel parts

Form of steel part	k_F
Plate	0.1013
Cylinder	0.0432
Sphere	0.0253
Cube	0.0338
Cylinder Z = 0.5 D	0.0639
Cylinder Z = D	0.0303
Cylinder Z = 2 D	0.0391

TABLE IV
Thermal diffusivity of selected metals in m^2/s [21].

Material	a , m^2/s
Silver (99.9 %)	1.6563×10^{-4}
Copper at 25°C	1.11×10^{-4}
Aluminum	9.7×10^{-5}
AISI 1010 steel	1.88×10^{-5}
Steel containing 0.5% carbon	1.48×10^{-5}
Steel containing 1% carbon	1.172×10^{-5}

During transient nucleate boiling process the surface temperature of steel part maintains at the level of boiling point of a liquid and insignificantly differs from it [22, 23]. The last characteristic was called self – regulated thermal process (see Fig. 9 and Fig.10) [7, 22, 23].

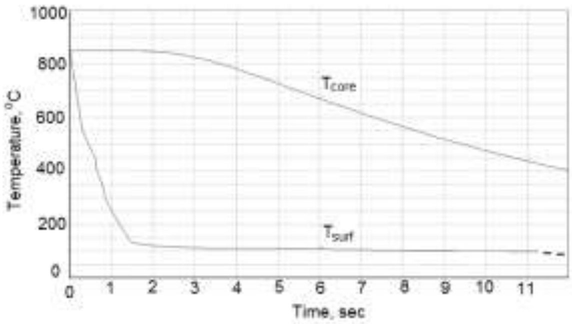


Fig. 9: Cooling curves obtained in Idemitsu Kosan Co., Ltd. lab, (Japan) for cylindrical specimen of 28 mm diameter and 112 mm length when quenching in water flow of 1.5 m/s at 20°C [22]

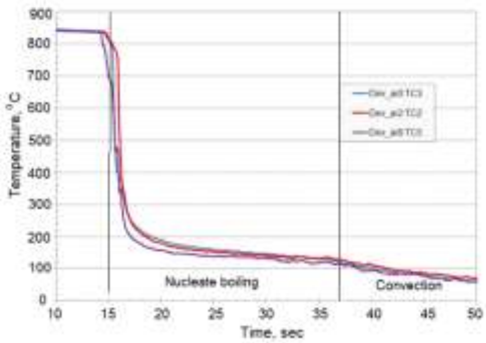


Fig. 10: Cooling temperature curves measured very close to surface during quenching cylindrical 2 inch probe in agitated cold water [23].

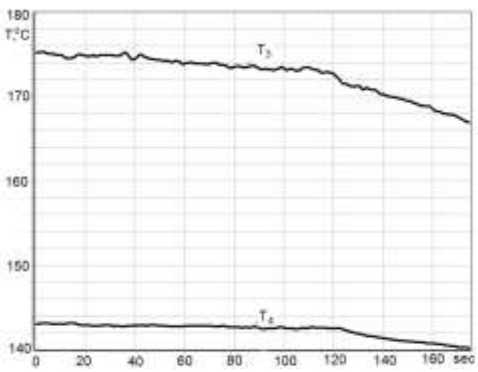


Fig. 11: Surface cooling curves at the distance of 220 mm and 290 mm from the end of the Probe [16]: T3 is thermocouple 3, T4 is thermocouple 4.

Fig. 11 presents temperatures curves recorded by thermocouples 3 and 4 installed at the not heated end of the probe showing the transition from the nucleate boiling process to convection at a time 120 sec. As seen from these data, the difference in time when this transition took place is very small between these two points. It can be explained by a very high speed of the thermal wave movement. The distance between thermocouple 3 and thermocouple 4 is 0.07m, and the speed of the thermal wave movement, according to authors [6, 14] is rather high. It is not clearly understandable why there is a small difference in time transition from nucleate boiling process to convection recorded by thermocouples T_3 and T_4. It will be possible to answer a question after solving hyperbolic heat conductivity equation taking into account in the boundary condition the effect of the double electrical layer. Meanwhile, the example how one can evaluate the average thermal diffusivity of steel is provided below.

Example: Using experimental data shown in Fig. 9, calculate average thermal diffusivity of steel if diameter of cylindrical long sample is 28 mm and water flow is 1.5 m/s. According to authors [11], water flow 1.5 m/s provides convective heat transfer coefficient equal to 6900 W/m²K. Convective Biot number for sample 28 mm is $Bi = \frac{6900W/m^2K}{22W/mK} \times 0{,}014m = 4.393$. According to Fig. 8, the value Ω is equal to 1.65. Using equation (14), we evaluate average thermal diffusivity as:

$$a = \overline{\Omega} k_F \frac{D^2}{\tau_{nb}} \qquad (15)$$

or

$$a = 1.65 \times 0.0432 \times \frac{0.000784m^2}{11.1s} = 5.035 \times 10^{-6} m^2/s.$$

Obtained result coincides very well with the average thermal diffusivity of stainless steel AISI 304 used for interval temperature 20°C – 850°C.

VI. PHYSICAL PROPERTIES OF MATERIALS EVALUATION

As one can see from Fig. 12, the oscillation of surface temperature of massive cylindrical steel probe takes place versus time.

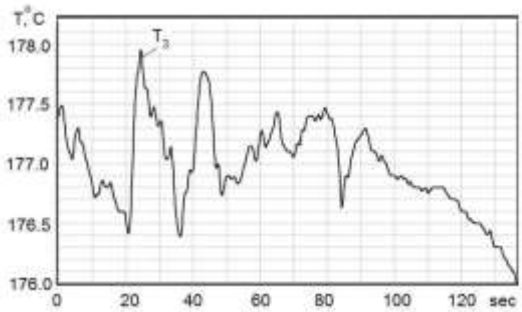


Fig. 12: Oscillation of the surface temperature at the distance of 220 mm (thermocouple 3) when cooling the probe in the 12% water solution of NaCl at 20°C

The thermocouple 3 was accurately welded to the metal surface and it was all time in air not in liquid and probe was fixed not agitated. One reason is left to explain observed oscillation. At the bottom of cylindrical probe, where double electrical layer was formed (see Fig. 6 and Fig. 13), oscillating local film boiling bubbles appeared that was recorded by free electrons reflected by double electrical layer. Due to huge thermal inertia, surface temperature of probe cannot oscillate. Such oscillation can produce only free electrons which were reflected by double electrical layer.

In all experiments, the surface temperature (recorded by the thermocouples located at areas of the probe (which were not immersed into the electrolyte) was oscillating as shown in Fig. 12. A question is arising regarding the nature of the observed temperature oscillations since the above thermocouples were in the still air, the probe was properly fixed and the thermocouples were properly welded to the probe surface. Since the probe was fixed after its immersion into the electrolyte and the electrolyte was not agitated, a local film boiling could appear at the bottom of the probe. These local vapor bubbles are usually unstable and could affect significantly the double electric layer (see Fig. 13). Bubble oscillation was recorded by welded thermocouple due to free electrons in steel [25]. If further investigated, the signals created by free electrons in metal can be used for the physical properties of metals evaluation.

VII. DISCUSSION

To understand better the poker effect, let's consider some well-known facts of the statistical physics [26]. Proceeding from the classical statistical thermodynamics, it is possible to explain thermal waves as the movement of free electrons. According to the statistical thermodynamics, the average kinetic energy E of electrons is [26]:

$$\overline{E} = \frac{3}{2}kT \qquad (16)$$

On the other hand, the movement of free electrons in metal creates very high pressure which can be calculated by equation (17):

$$P = \frac{2}{3}n\overline{E} \qquad (17)$$

Here n is a number of electrons in one sm^3 of metal; k is the Boltzmann constant which is equal to $k = 1{,}3806488(13)\times 10^{-23}[K^{-1}]$ [26]. As it follows from equations (16) and (17):

$$P = nkT \qquad (18)$$

It means that the pressure created by free electrons in metal is directly proportional to the absolute temperature T.

Fig. 13 shows the movement of free electrons in cylindrical probe due to created pressure (18).

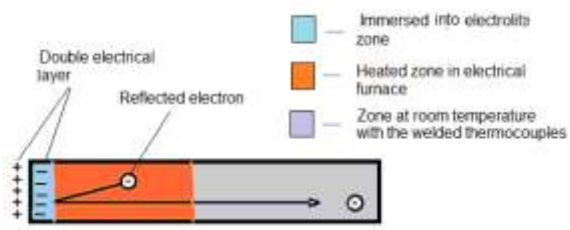


Fig. 13: Schematic that explains a poker effect based on laws of statistical physics [14].

Unfortunately, after painstaking experimental investigation of initial phase of quench process made by French in 1930, there was a time when scientists used only one thermocouple instrumented at the center of the probe. Also, considering effective heat transfer coefficients, engineers made a incorrect conclusion on possibility of measuring surface temperature by central thermocouple due to $T_{center} \approx T_{surface}$ as they thought was true for standard cylindrical probe 12.5 mm in diameter [11] when calculating Biot number $Bi \le 0.2$. Such incorrectness was widely discussed in the paper [27]. The paper shows that careful investigation of initial quench process opens a great possibility for physical properties of materials accurate measurement. Mathematician and physician should continue solving this problem on the basis of hyperbolic heat conductivity equation, accurate experiments which, as seen from the paper, is very important for the practice [28, 29]. Developed new technologies [19, 30] can be implemented widely in heat treating industry if they are supported by appropriate calculations, software which require of knowledge of physical properties of materials. Also, it is recommended to pay more attention to study the physics of the initial quenching processes which, unfortunately, are not deeply investigated yet.

VIII. CONCLUSION

New methods of evaluating physical properties of materials (thermal diffusivity a (m^2/s) and relaxation time τ_r (s)) are proposed based on duration of transient nucleate boiling process and speed of thermal wave distribution measurement. The measured values a and τ_r are used for the quench process recipes development and prediction the film boiling mode when immersing heated to high temperature steel parts into liquid during quenching. Measured by such a way thermal diffusivity of material is already used in heat treating industry while relaxation time τ_r (s) requires additional testing to weigh up the accuracy of the performed measurements.

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