Analysis of acoustic emission effect accompanying metal crystallization

V B Vorontsov\textsuperscript{1,3} and V V Katalnikov\textsuperscript{2}

\textsuperscript{1}Urals State University of Railway Transport (USURT), 66 Kolmogorov Street, Ekaterinburg, 620034, Russia,
\textsuperscript{2}Ural State Technical University (USTU), 19 Mira St, Ekaterinburg, 620002, Russia,

E-mail: vorontsov@bgd.usart.ru, vdeka@mail.ru

Abstract. The work is devoted to acoustic emission (AE) which occurs in the process of crystallization of metals and non-organic substances. The objective of the research is to obtain an AE signal from the interphase surface and carry out frequency analysis of the spectrum. The results of the research have shown that the signals generated have maximum amplitude in 21-27 kHz range with maximums at 21 and 24 kHz at $v=0.8$ cm/sec and at 21 and 23 kHz at $v=1.43$ cm/sec. To explain the results obtained the cluster model of melts was applied. Every AE signal corresponds to the advance of interphase surface by a step equal to 3 layers of clusters.

Introduction

Acoustic emission is the emission of sound waves in sound and ultrasound band by materials, which undergo inner reconstruction and release elastic energy. The acoustic emission (AE) effect was more then once observed and is thoroughly studied in solids at phase transformations and is almost unexplored at crystallization and melting. Moreover, at 1-st order phase transformations there is the dynamic restructuring which can also generate sound signal if the transformation goes impulsively and is connected with elastic properties of the reconstructing phases.

This was experimentally proved in 1957 by Joseph Kaiser \cite{1,2}, who was the first to detect AE at solidification of Sn, Zn, Pb, Sb metals. In spite of the fact that AE was observed many times at crystallization of metals and alloys by Borchers and Tenzy \cite{3-5} and in nonorganic substances by Shokarov \cite{6}, there is still no satisfactory explanation of this phenomenon.

The researchers suppose that there is connection between AE and contraction which most of the metals undergo when solidify or in case of anomalous metals Sb and Bi melt.

There is actually a different point of view expressed by Tensi and Radtke \cite{5}, who studied crystallization of Pb-Sn and Cd-Zn, that the emission is caused by friction of neighbor grains and clashing dendrites, which grow to meet each other.

This statement can be easily checked through experiment when the conditions for the growth of single crystal with plane solidifying interface are created because in this case the sources of emission proposed by the authors are excluded.

This topic is now thoroughly investigated because of the strong practical interest to the AE effect. The scientific interest is explained by the possibility to use AE effect as a sensitive method to study the processes of crystallization. From the practical point of view, there is the opportunity to influence the melt by the sound in resonance with the frequencies generated at crystallization and thus affect the casting structure.

\textsuperscript{3} To whom any correspondence should be addressed.
The objective of this research is to identify the source of acoustic emission in ultrasound frequency band.

**Experimental part**

Aluminium (with 99.999 purity) was chosen as a model material. To exclude any influence of grains’ boundaries on the AE a special method was developed which allowed to observe and analyze acoustic signals in the process of single crystals growth. The developed device (figure 1) permitted to get single crystals of metals with melting point up to 1000 centigrade degrees in the atmosphere of spectrally pure helium. The single crystals of aluminium were grown using the Bridgeman method. The seed crystals for aluminium with <100> orientation in a soft crucible made from boron nitride (BN) were used.

The experiments on single crystals grow were organized as to exclude possible sources of emission, which are not connected with the processes on the interphase surface (IS). The method allows to get a thermogram of cooling and to analyze the acoustic spectrum of AE signals while single crystals were growing.

In the experiment, the content of the crucible was melted and then the alundum waveguide 3 (figure 1) was put into the melt so that the distance between the seed crystal and the waveguide was 15mm. The move up of the furnace was then switched on, this provided the crystal growth with immobile crucible.

![Figure 1](image.png)

**Figure 1.** Experimental installation for AE study at metal crystallization. 1 – heating furnace, 2 – crucible with melt, 3 – piezoconverter with waveguide and thermoelement, 4 – 4 – high-frequency temperature regulator BPT-2, 5 – temperature recorder KCI-4, 6 – AF-15 device, 7 – oscilloscope C8-13, 8 – digital oscilloscope ACK-3106, 9 – PC, 10 – heat shield

In the experiments two speeds were set – 0.8cm/min and 1.4cm/min with thermal gradients of $G_1=12 \text{ grad/cm and } G_2=25 \text{ grad/cm}$ along the crucible’s axis.

The present research work is devoted to the study of AE at crystallization in ultrasound part of spectrum. To achieve the objective, the device 7 (figure 1) was re-equipped so that the whole information, acquired during the experiment, was transmitted to a PC. For this purpose, the PC-based digital storage oscilloscope ACK-3106 was used (figure 1). The software used allows to carry out real time spectrum analysis of AE signals in the 10-50 kHz band.

The analog signal after its amplification by the acoustic emission device AF-15 (6) went to the digital oscilloscope, connected with the PC by the signal cable. The software of ACK-3106 made it
possible to convert analog signal into digital one, perform necessary processing of it and carry out Fourier analysis.

\textbf{Figure 2.} 3D image of spectrum amplitude-frequency analysis, obtained while growing crystals at 0.8 cm/min speed. It follows that AE signals appear at the start of crystallization, but their repetition rate is not constant. The highest density of the signals is at the beginning of growth and 40 seconds before the end of solidification process which in our case is 200 seconds in total.

\textbf{Figure 3.} The diagrams of events' frequency distribution with frequency intervals of 500Hz and it follows that 90% of all events is in the frequency interval of 21-27 kHz. More detailed analysis within the intervals showed that the events with 21 kHz and 24 kHz frequency are realized, but among these two the former appears 2.6 times more often.

Such situation repeated for single crystals which were got at 7.4 cm/min speed. The 21 kHz frequency was realized, the second frequency was very close to it – 22.5 kHz.

On the basis of the spectra analysis it can be suggested that the frequencies observed correspond to two aspects of the normal growth mechanism realization. The first frequency corresponds to the formation of 2D nucleus, and the second frequency corresponds to the standard layer motion of the solidifying surface. After the layer is formed, the IS moves to the next stage (layer). When the speed of IS advance grows, the kinetics of the process changes and this affects the results of analysis – the frequencies rise also.
Theoretical part

As AE is closely connected with the process of crystallization, in order to understand the former it is necessary to realize possible mechanisms of the latter.

First of all the observation of Ubbelohde must be taken into account that at the crystallization speeds considered (\(v > 10^{-2}\) cm/s) the conversion into the crystalline state of the single atoms is impossible. Therefore we suggest that a crystal layer is formed by clusters which in the Aluminium melt have size of approximately 21 Å, or by blocks of another type, containing bigger number of particles.

AE occurrence can be understood in the following way. From the quantum-mechanical point of view the process of crystallization can be represented as the transfer of excess energy \(q_{12}\) to a crystal by every settling particle. This energy mainly turns into heat energy, in other words into the energy of additional backgrounds, scattered in the environment due to heat conductivity:

\[
\left|i, \{n_q\}\right| \rightarrow \left|f, \{n_q'\}\right|.
\]

Here \(i\) and \(f\) - the initial and the finite state of crystal electronic subsystem at a single settling, and the set of quantum numbers \(\{n_q\}, \{n_q'\}\) characterizes its background subsystem.

At the same time, the heat transfer without excitation of the background subsystem is possible:

\[
\left|i, \{n_q\}\right| \rightarrow \left|f, \{n_q\}, \bar{p}\right|,
\]

\(\bar{p}\) – mechanical impulse, transferred to a whole crystal. This energy transfers into the sound oscillations of a whole crystal, which can be detected by the devices.

The number of impulses, equal to the number of settling particles at a time unit, multiplied by the Debye-Waller factor, constitutes the primary carrier frequency. At the temperature higher then the Debye temperature, the Debye-Waller factor is equal to (according to [3]):

\[
\exp \left\{ -\frac{3R}{2k_b \Theta} \left[1 + 4\pi \frac{\Theta}{\Theta_D}\right] \right\}, \quad \Theta \geq \Theta_D
\]

\(\Theta\) - absolute temperature, \(\Theta_D\) – the Debye temperature, \(R\) – the recoil energy, got by a particle at the moment of conversion. Here, the rectangular sign means >.

The melting heat for the atoms of aluminium is 0,1 electron volts, consequently, the recoil energy \(R\) will be about atomic \(q_{12}\), multiplied by the number of atoms, participating in a single act of conversion.

The existence of two mechanisms of scattering (1) and (2) explains random distribution of the acoustic signal, which is observed during the experiment (figure 2). At the same time it must be considered that the sound wave pressure can occur only if specific volume changes and it should be taken into account in calculations.

In addition to this microscopic picture, the following considerations can be expressed.

Close to the point of phase transition, the system is in a non-equilibrium state so the role of thermodynamic values fluctuations is growing. According to (10), fluctuation of volume \(\langle V \rangle\) of a body’s certain part with a certain number of particles \(N\), and respective fluctuation of particles' quantity are related through the following ratio:

\[
\frac{(\Delta N)^2}{N^2} = \frac{\langle \Delta V^2 \rangle}{\langle V^2 \rangle}
\]

We should also consider Frenkel's reasoning [5] that the melt close to the point of the phase transition is in almost ordered state and it contains some vacancy hollows. Their volume is approximately equal to excess volume of the melt comparing to the volume of the crystal. The size of the hollows, apparently, is smaller then the critical one (11), which is \(10^{-5}\) for aluminium, and therefore they collapse instantly, making a crystal to sound. The acoustic sound is the envelope of pulses which
number changes in the process of layer formation. This is the explanation for the periodicity of the signal.

Now quantitative estimations should be given.

One-dimensional model (when the interphase boundary (IB) is plain and perpendicular to Ox axis) is examined. Temperature gradient is located along the axis; this provides banded crystallization without grains. Let \( N(t) \) is the number of particles which moved from melt to crystal state during a time unit. The change of quantity in the line of Ox is step-wise but taking a microscopically small period of time so that the change can be considered as continuous.

To calculate \( N(t) \) it is necessary to identify time dependencies of particles quantity distribution in a base layer for \( n \) particles in a base layer and for \( m \) particles in a neighbor layer. The latter define the roughness of crystallization IB.

We introduce local time \( t \) which characterizes the process of particles accumulation in a base layer:

\[
N(t + \tau) = \sum_{n=0}^{N} n \cdot p(n, \tau), \quad N(t + T) = N(t)
\]

where \( p(n, \tau) \) - probability of \( n \) particles settling at \( \tau \) time, \( T \) - time of one layer formation.

We consider that completion of a layer by particles is the Poisson process. So the probability of a single particle settling can be determined with the following equations:

\[
p(n, \tau + d\tau) = p(n, \tau) \cdot p(1, \tau),
\]

\[
p(0, \tau + d\tau) = p(0, \tau)(1 - p_1),
\]

where \( p_1 = n_0 d\tau \) - probability of a single particle settling at \( d\tau \) time, \( n_0 \) - average number of particles, settled at a time unit. The probability of particles’ accumulation in a neighbor layer is defined by the same equations (6), but with a different settling velocity \( m_0 \neq n_0 \). So the solution for the last equation (6) is the Poisson distribution:

\[
p_n(m, \tau) = \left(\frac{m_0 \tau}{m!}\right)^m e^{-m_0 \tau}
\]

and average number of particles \( \tilde{m} \), settled in a neighbor layer at the time of base layer formation \( T \), becomes a seed crystal for a new layer formation.

Now we need to derive the equation for \( N(\tau) \) function (we temporarily omit part of the argument \( t \)). The velocity of particles’ accumulation in a base layer is, evidently, proportional to the velocity of their settling \( n_0 \), number of particles settled \( N(\tau) \) and vacant sites \( N - N(\tau) \), where \( N \) - total quantity of particles, forming a layer:

\[
\frac{dN(\tau)}{dT} = n_0 N(\tau)(N - N(\tau))
\]

Boundary condition can stated in the following way:

\[
N\left(\frac{T}{2}\right) = \frac{N}{2},
\]

where \( T \) - time of layer formation. The solution for equations (8), (9) looks like:

\[
N(\tau) = \frac{N}{1 + \exp\left(-Nn_0\left(\tau - \frac{T}{2}\right)\right)},
\]

This conception doesn’t give fully explanation to observed peculiar features of spectrum, but allows to evaluate the order of the frequencies detected. If we take that every layer has thickness of 21 Å, for 0.8 cm/min speed the frequency will equal
\[ v_T = \frac{1}{T} = \frac{v}{l} \approx 62 \text{ kHz}, \]

for 1.4 cm/min speed \( v \approx 110 \text{ kHz}. \)

Experimentally we observed the frequencies which are three times lower.

On the basis of the research it can be said that the growth of Aluminium single crystals goes with acoustic emission signals in 21-24 kHz band. In further theoretical investigation of the subject, we proceeded from the cluster model of the melt and normal mechanism of crystallization. This allowed to conclude that AE signals are caused by the collapse of vacancies formed in the process of clusters’ settling on the IS during crystallization.

Quantitative assessment of experimental results with theoretical calculations showed that the occurrence of AE signals is strictly regulated by frequency and one signal correlates with the settlement of 3-5 layers of clusters that means the advance of the solid phase for the thickness of this layer.

References
[1] Kaiser J 1957 Forching-Wes. 23, 38
[2] Borchers H and Kaiser J 1958 Akustische Effekte bei Phasenumbergangen in System Blei-Zinn.-Z. Metallkunde, 2, 49, 95-100
[3] Tensi H 1974 Acoustic Emission Measurements During Crystallization and Melting of Metals and Binary Alloys (Tokyo: Japan Industrial Planning Association) 46, 1974.
[4] Borchers H and Tensi H 1963 Piezoelektrische Impulsmessungen bei der Phasenänderung von Unlegierten Proben der Binaren Legierungen Blei-Zinn, Blei-Antinom und Kadmium-Zink (Metall) 8, 17, 784-7
[5] Tensi H and Radtke W 1978 Schallemissionmessungen bei Phasenänderungen (Metall) 32, 681-5
[6] Zadumkin SN, Hokonov HB and Shokarov HB 1975 JETF 4, 68, 1315-20
[7] Vorontsov VB and Bezdomov AK 1988 Acoustic Effect and Crystallization of Aluminium (Moskva: Institute of crystallography AN USSR), 32-34
[8] Ubbelode AR 1982 Rasplavlennoe Sostojanie Veshchestva (Moskva: Metallurgiya)
[9] Kittel Ch 1967 Kvantovaja Teorija Tverdih Tel (Moskva: Nauka)
[10] Landau LD and Livshic EM 1964 Staticheskaya Fizika (Moskva)
[11] Frenkel YI 1972 Vedenije v Teoriju Metallov (Moskva: Nauka)