DATA ON THE TECHNOLOGICAL CHANGE OF COPPER ALLOYING IN THE LIGHT OF FINDS EXCAVATED AT THE TUMULUS OF REGÖLY•

ADATOK A RÉZ ÖTVÖZÉSÉNEK TECHNOLÓGIAVÁLTÁSÁHOZ A REGÖLYI TUMULUS LELETEINEK TÜKRÉBEN

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Abstract

We observed a Sn-Ni phase unnoticed earlier during the archaeometallurgical examinations even in several of the bronze objects of the tumulus of Regöly that can be linked to the early Ha D_1 period, showing substantial changes in its finds and traceable via Asia Minor to Central Asia in its interrelations. In examining and revealing the reasons for the unique phenomenon, focus was being shifted increasingly to the raw material, the way copper had been alloyed. The fundamental step of bronze workmanship in the prehistoric age that is alloying copper with tin still triggers a number of disputes. Making tin bronze has basically four options that we know: melting a mixture of metallic tin and copper; adding naturally occurring tin oxide (tin ore or cassiterite, SnO_2) to already molten copper; smelting an ore containing both copper and tin; reducing a mixture of copper ores and tin ores. Based on archaeological field observations, it is an overwhelming opinion on our continent that alloying to the raw material for the finds of the European Bronze Age was performed still in the smelting phase using a mixture of ores of the proper composition. At the same time, successful archaeological experiments demonstrate that it was possible to directly alloy pure metals, copper and tin, under the technical circumstances of the age in question. However, Europe shows no sign of having as intense production at significant archaeological sites of tin ores as at copper mines of the prehistoric age. Moreover, we are not aware of commerce of tin of any significant volume; the use of objects made from pure tin did not become widespread up to the end of the Bronze Age anywhere on our continent. The few tin objects known seem to be extremely low compared to the quantity of bronze produced in that age for the case of alloying pure copper even if we assume the melting of the majority of tin objects. Also, no proof of alloying pure-metallic tin has been shown as yet at archaeometallurgical sites in the area of Europe during archaeometallurgical examinations, to our knowledge. The data referring to the direct alloying of copper unknown earlier in the European material of the new Sn-Ni phase observed in the Regöly finds examined recently also proves that manufacturisation and the accompanying explosion-like technological advance is not the outcome of the internal development of the Hallstatt culture sphere. It had been a ready-implemented practice in Europe by masters of the last Ionian migration starting from Asia Minor in the last third of the 7th century BC in the Carpathian Basin just as had been the technology of alloying copper directly with metallic tin.

Kivonat

A Ha D1 időszak kezdetéhez köthető, leleteiben gyökeres változásokat mutató, kapcsolatrendszerében Kis-Ázsián át Közép-Ázsiáig követhető regölyi tumulus bronztárgyainak archeometallurgiai vizsgálata során több tárgynál is egy korábban nem tapasztalt Sn-Ni fázist figyeltünk meg. A különös jelenség okainak vizsgálata és feltárása során egyre inkább az alapanyag, a réz ötvözésének módjára terelődött a figyelem. Az őskori bronzművesség alapvető lépése, a réz ónnal való ötvözésének módja még ma is sok vitát vált ki. Az ónbronzok előállításának alapvetően négy lehetősége ismert: fémes ón és réz keverékének megolvasztása; a természetben előforduló ón-oxid (ónkő vagy kassziterit, SnO_2) hozzáadása a már olvadt rézhez; rezet és ónt együttesen tartalmazó érc

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kohósítása; réz- és ónércek keverékének redukciója. A terepi régészeti megfigyelések alapján kontinensünkön az az általános vélemény, hogy az európai bronzkor leleteinek nyersanyagához az ötvözést még a kohósításkor, a megfelelő összetételű ércek keverékének felhasználásával végezték. Ugyanakkor eredményes régészeti kísérletek mutatnak arra, hogy az adott kor technikai lehetőségei mellett is lehetséges volt a színfémek, a réz és ón direkt ötvözése. Az ónércek jelentős lelőhelyein azonban Európában nincs nyoma az olyan intenzitású kitermelésnek, mint az őskori rézbányáknál. Ráadásul jelentős mennyiségű ón kereskedelmére utaló leleteket sem ismerünk; kontinensünkön a tiszta ónból készített tárgyak használata a bronzkor végéig sehol sem vált általánossá. A néhány ismert ón tárgy a korszakban gyártott bronz mennyiségéhez képest a tiszta réz ötvözése esetén rendkívül kevésnek tűnik még akkor is, ha feltételezzük az ón tárgyak döntő többségének a beolvasztását. Ráadásul az eddigi archeometallurgiai vizsgálatok során tudomásunk szerint Európa területén az őskori lelőhelyeken még nem találtak bizonyítékot a fémtiszta ónnal való ötvözésre. A most vizsgált regölyi leleteknél megfigyelt új Sn-Ni fázis az európai anyagban korábbról eddig ismeretlen, a réz direkt ötvözésére utaló adata is megerősíti, hogy a manufakturalizálódás, és az azzal együtt járó robbanásszerű technológiai fejlődés nem a hallstatti kultúrkör belső fejlődésének eredménye. Az a Kárpát-medencébe a Kr. e. 7. század utolsó harmadában a Kis-Ázsiából kiinduló utolsó ión vándorlás mesterei által Európába készen átültetett gyakorlat, miként a réz direkt módon, fémes ónnal való ötvözésének technológiája is.

KEYWORDS: CAULDRON, CHAIN, INCENSER, COPPER, TIN, NICKEL, LEAD, METAL-TO-METAL ALLOYING, MANUFACTURISATION, SERIAL PRODUCTION

KULCSSZAVAK: BOGRÁCS, FÜSTÖLŐ LÁNC, RÉZ, ÓN, NIKKEL, ÓLOM, FÉM-A-FÉMMEL ÖTVÖZÉS, MANUFAKTURIALIZÁLÓDÁS, SOROZATGYÁRTÁS

Introduction, archaeological background

The fundamental step of bronze workmanship in the prehistoric age that is alloying copper with tin still triggers a number of disputes. Making tin bronze has basically four options that we know: melting a mixture of metallic tin and copper; adding naturally occurring tin oxide (tin ore or cassiterite, SnO_2) to already molten copper; smelting an ore containing both copper and tin; melting a mixture of copper ores and tin ores (Heeb & Ottaway 2014, 178). Based on archaeological field observations, it is an overwhelming opinion on our continent that alloying to the raw material for the finds of the European Bronze Age was performed still in the smelting phase using a mixture of ores of the proper composition (Hauptmann 2014, 94). At the same time, successful archaeological experiments demonstrate that it was possible to directly alloy pure metals, copper and tin, under the technical circumstances of the age in question (Heeb & Ottaway 2014). This is also confirmed by the tin ingots found in the Uluburun and Salcombe ship finds, based on which the force of the question of tin commerce necessary for bronze production arises as well (Harding 2005; Harding 2013, 375). However, Europe shows no sign of having as intense production at significant archaeological sites of tin ores as at copper mines of the prehistoric age (Wang et al. 2016, 80). Moreover, we are not aware of commerce of tin of any significant volume; the use of objects made from pure tin did not become widespread until the end of the Bronze Age anywhere on our continent. Views without proper foundation are not considered in this study, such as tin pest causing the disappearance of a significant portion of Bronze Age ingots. Based on our results from the examinations we specifically looked for parallelism in the direction of ore deposits with high nickel content. Their relative concentration has so far been observed on the territory of Switzerland alone where Margarita Primas collected Bronze Age tin finds that she dated them back to the late Bronze Age, the centuries 11th - 10th BC (Primas 1984). These objects mostly classified as small jewels have lately been supplemented by a tin ingot found at the Sursee–Gammainseli archaeological site (Nielsen 2014). However, these couple of objects seem to be extremely low compared to the quantity of bronze produced in that age for the case of alloying pure copper even if we assume the melting of the majority of tin objects. Also, no proof of alloying pure-metallic tin has been shown yet at archaeological prehistoric sites of Europe during archaeometallurgical examinations, to our knowledge. Therefore, we handled the phenomena referring to the alloying in the age in question with priority when examining the samples from the mound of Regöly (Szabó & Fekete 2011; Fekete & Szabó 2017a).

The surroundings of the finds, the archaeological site at Regöly

The archaeological site is located in the Western half of the Carpathian Basin, in the Southern part of Transdanubia, at the confluence of the Kapos and Koppány rivers (**Fig. 1.**). This site was a place of excavating of a tumulus in 2011 to 2012 whose material dates clearly back to the last third of the 7th century BC, the Iron Age, yet, but cannot match either the finds of the Scythian or that of the Hallstatt culture (Szabó & Fekete 2011, 49; Fekete & Szabó 2015).

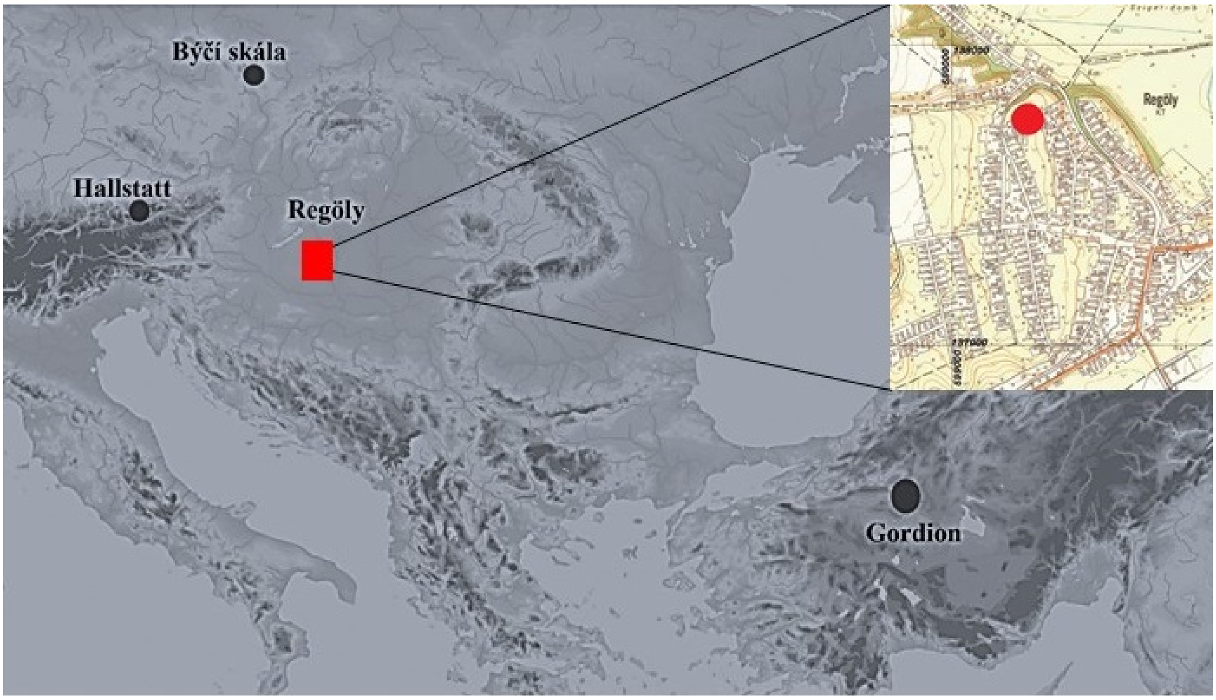

Fig. 1.: The location of Regöly in the Carpathian Basin. Regöly is in the Southern part of Transdanubia, at the confluence of the Kapos and Koppány rivers

1. ábra: Regöly elhelyezkedése a Kárpát-Medencében. Regöly a Dunántúl déli részén fekszik a Kapos és Koppány folyók összefolyásánál

Thus, it is of utmost importance even from this aspect that two contradicting positions have been established by the European and the Russian research in connection with the emergence of the Hallstatt culture. On the European side, according to the majority, the continuous internal development, transformation ongoing even after the late Bronze Age had been complemented by the commerce of Eastern objects and cultural influence (Metzner-Nebelsick 2002). On the part of Russian researchers, I. V. Bruyako, the appearance of Eastern horse harnesses and weapons in the West can be explained by mass migrations only (Bruyako 2005). As indicated by the latest finds at Regöly, however, an intermediary solution arises: the replacement of the ruling elite seems to be the most probable (Szabó & Fekete 2011, 49–50; Fekete & Szabó 2015; 2017b; Szabó 2015; Hansen 2011; 2017).

Those who excavated at the mound of Regöly, the objects revealed can be the legacy of the people called Sigynnii (sigynnoi) or later Pannoni who ruled the area of South Transdanubia southward from Lake Balaton from the second half of the 7th century to the beginning of the 4th century BC, almost 3 centuries (Szabó & Fekete 2011, 48-50). Objects or appearances orientating towards Asia Minor unknown so far on our territory can also be observed in the material of the mound showing close relations to the Hallstatt finds (Fekete & Szabó 2015; Szabó et al. 2018). The mound structure itself built from stamped clay exhibits the utmost parallelism to one located in Gordion, Phrygia (Young 1981, 84. Fig. 52;Fekete & Szabó 2017 Fig. 8).We encountered a rare and special mineral (Cr-spinel) in the ceramic during the archaeometric examinations, which is unknown in the area of Europe but can also be found in the control sample from Gordion (Gyöngyösi et al. 2017; Kürthy et al. 2018). At the same time, the three-feathered bronze arrowheads, reptilian scale armour sheets (Horváth & Szabó 2015), the harnesses oriented towards Inner Asia (Szabó & Horváth 2016), the ceramic fragments, crockeries to be linked to both the Andronovo culture and the Ancient East (Kürthy et al. 2016) reveal, too, that a nation was reckoned with to be of Eastern roots who were, though, similar to Scythes of the steppe in their way of life but had bypassed the Black Sea not from the North but from the South, Asia Minor. Based on our current knowledge there was one and only such a nation in the period examined, the Cimmerians indicated in ancient sources who, in part, bypassed the Black Sea from North fleeing from the Scythes, while their other branch passing via the Caucasus fought on the territory of Urartu, Phrygia, and later Lydia already from the 8th century BC (Bruyako 2005, Fig. 31; Kohler 1995, 185–189). This is how the unique blend of objects of Eastern horse people and the advanced technology of the Ancient East becomes understandable and interpretable in the legacy of those arriving from the former Median Empire to South Transdanubia via the Balkans in the last third of the 7th century BC according to ancient sources (Szabó & Czuppon 2014, 50–51. Fig. 1).As a result, tracing the historic background and connection system is exceptionally important to understand the complex data of these objects, showing radical technological changes in their forms and manufacturing methods due to the strong mixing of different traditions. It is considered crucial that the archaeometrical results of each object are carefully examined and evaluated together with the continuously developing historical and social connections before the disclosure of the full material on the excavation of Regöly.

When observing a part of the metal objects in a preliminary manner, we remarked technological phenomena that had gone undetected so far that is they refer to serial production and special technologies assembled from composite elements (Szabó 2013; Fekete & Szabó 2017a). We complemented the usual search for shape similarities and the determination of material composition of these archaeological objects by detailed metallographical examinations. We selected objects for these examinations that had been produced in a serial manner and their making had required special know-how of technology and a well-equipped workshop. Below we present the examination results of two object types closely linked to sacred activities from the line that is a cauldron used for offerings of food and drink as well as an incenser chain. These exhibited results referring to the way of alloying unknown for the objects of the Carpathian Basin so far.

Preliminary results, examination of the cauldron found at Regöly in 1907

Up to now, a relatively intact bronze cauldron (**Fig. 2.**) was earlier excavated from Regöly, and fragments of at least another bronze cauldron has lately been excavated from the tumulus. The first one, fitted with doubled cross-shaped slings and pseudo-twisted nibs, slightly oval, with a flange diameter of 26 and 27.3 cm, cauldron with a missing bottom reached the museum of Szekszárd under unknown circumstances in 1907 (Patay 1990, 30). It can be well seen that two parallel lines run in two bands each under the flange on the external side on the old Regöly cauldron (Szabó 2009; 2012; 2013). The line design of the ornamentation is continuous, it seems to be carved. On the flange of a thickness of about 3 mm, in the proximity of the slings mounted with 4 cast-looking lens-shaped rivets on both sides, 2 slightly thickening parts protruding at 1.5 cm from the flange can be observed that shows uneven breakage surface in contrast to the smooth surface of the edge of the flange. Except for the corrosion phenomena, the external and internal sides of the cauldron is smooth in the upper third, whereas its side decreases to foil-thin towards its bottom. Signs of forging can be seen on both of its sides. (A considerable part of its bottom was missing that was complemented by restorer Mihály Nacsa using artificial resin.) We took sample from the part near the sling of the flange of the cauldron. The sample preparation and instrumental analysis of sample no. BrWMM63 were performed by support of G. McDonnell and M. Pollard, Head of Division at the Division of Archaeometallurgy. Embedded cold in resin and ground wet, we etched the carefully polished cross-section using an $FeCl_3$ solution.

Fig. 2.:

The intact bronze cauldron found at Regöly (1907)

2. ábra:
A Regölyben talált (1907) ép bronz bogrács

Almost nothing can be seen at low magnification on the cross-section image but the boundary of relatively large crystallites in the alpha structure is already silhouetted well at a magnification of 400, among them the twin-crystallites split along the straight limiting lines, hidden in some places. The cross-sectional image reveals copper sulphide inclusions in the spots located in a dispersed way, the distribution of tin is even, with a ratio of 8.89%. The annealed grain structure seen in the cross-sectional image with the twin-boundaries observed in some places refers to the fact that the object had been heat-treated after its plastic deformation. As few twin-boundaries can be observed, we can deduce from the grain structure that heat treatment had not been performed at high temperature and heat ramp-up had been slow and not shock-like. This results from the dimensions of the object as well. The dispersed appearance of the twin-crystallites refers in general more to the lattice stresses arising due to heat treatment than a raw material forged or carved during a post-processing. Considering the part referring to the place of mould pin, the enguss, which is protruding and slightly thickens, observed on the flange, as well as the forge marks polished surface and the composition, it can be determined that the than bronze workman made the cauldron from a semi-finished product of a raw material produced originally by casting and suitable for further processing (Szabó 2009, 348). Our earlier examinations also highlighted that the cauldron was made in a workshop where it was fitted with components produced in a serial manner (doubled cross-shaped sling, pseudo-twisted nibs) (Szabó 2012).

Metallographical examination of cauldron fragments and incenser chain links excavated in the Pannon tumulus at Regöly

During the excavation of the tumulus at Regöly (Kürthy et al. 2013), a half palm-sized flange fragment of a cauldron was dug out from the yellow loess layer marked RHQ20 containing many find fragments, between the burnt, stony layers of the chamber (Kürthy et al. 2013), that included the remainder of a rivet. (The layer mentioned contained several pseudo-twisted nib fragments, too, which, however, could have belonged to cysts.) Similarly, almost one and half dozens of intact and fragmented, more-or-less square-shaped chain links were excavated from this and the connected layers (**Fig. 3.**).

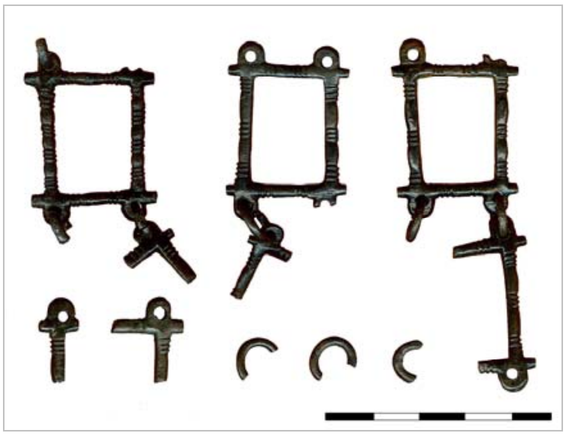

Fig. 3.: The excavated chain fragments from the Pannon tumulus of Regöly

3. ábra: A regölyi pannon sírhalomban feltárt lánctöredékek

Objects 8, 10, 14, 20, 22 and 45 of the fill-up of the tumulus at Regöly contained small-sized, cast, square-shaped, notched and formed from a stick-like shape, fragments of bronze chain (s) that were assembled from lugs fitted with 4 small nibs and rings bent from a bronze wire, posing apparently low-level technical challenge, unknown in the earlier finds material of the Carpathian Basin (Szabó & Fekete 2011, Table 20). Their function was clear to be determined at first sight: based on earlier, but of other design, of the same use, finds excavated also at Regöly (Fekete 1985b, Figure 1; 1986, Fig. 13). The appearance of incensers and the belonging chains can be regarded as almost massive in certain regions of the 7th – 6th centuries BC; parallel to a specific new cognitive-religious-ritual function (Fekete 2018).

The square-shaped cast chain links of the tumulus at Regöly exhibit many new technological processes. The jeweller removed all manufacture marks such that even the making is unclear whether it was through lost wax or some mould. That is, there are references uncertain and unclear to both procedures. There is a little burr at the nibs but it must be a worn-out, bulked-up part during use by the chain links. If it was a borehole, it had rather a mark only on the other side, but here it can be noticed on both sides of the hole. Small cross-directional notches can be seen in some places by the nibs. Such can be caused by gripping into a pair of pliers or a vice but they had had no such tools, as far as we know. It could be possibly a mark of carving or deformation but it seems to be completely unreasonable on such a small even surface where they appear.

Fig. 4.: Magnified photograph taken from a chain fragment. The photograph reveals some details of the processing technique and shows the ornamentation of the chain.

4. ábra: A lánctöredékekről készült nagy nagyítású fotó. A fotó felfedi a készítéstechnika pár részletét és a lánc díszítését.

One may think that these impressions may have possibly been generated by tiny differences of moulds. The linking rings had been clearly made using a wire processed by hammering. Their ornamentation is simple, small narrow 2 or 3 bean-pattern between groups of notches (**Fig. 4**).

The slightly trapezoid asymmetric shape of chain links of about 1.9 × 3.7 cm, the design of 4 nibs, the alternation of sophisticatedly divided beans and groups of notches are all workshop tricks that pop up not only in the direct neighbourhood (see Regöly hoard: Fekete 1995, Kemenczei 1996) but also in the bronze workmanship of the entire Transdanubia. In part (their elements) or in entirety, as copies, local version (s), including the metal working centres of paramount importance since centuries (Velem – Szentvid, Celldömölk – Ság-hegy, Keszthely – Apátdomb) where the new-style fibulas had been also made massively, as indicated by the semi-finished products and/or moulds (Fekete 1985b, Fig. 11-13; 1986, Fig. 8-10; 1995, 42., Fig. 1-5; 2006, 102., Table 61; Kemenczei 1996)

We selected a fragment of the cauldron from layer marked RHQ20 and that of an incenser chain link from the same object for complex archaeometallurgical examinations.

Examination methods

Description of sampling, the samples

During sampling, we cut off a slice to be examined in the full cross-section from the flange-side corner of the cauldron fragment (**Fig. 5.**).

Fig. 5.: The fragment of a cauldron from Regöly. The positions of the examined samples are signed in the photograph. The material of the cauldron and the rivet were examined.

5. ábra: A Regölyben feltárt bogrács töredék. A fotón a vizsgált minta helyét jelöltük. A bogrács és a szegecs anyagát vizsgáltuk.

We removed the rivet located on the sheet piece and embedded it fully, the flat polished specimen was made at the bottom of the rivet. This way, it was possible to examine its entire cross-section. In the same manner, we added one of the nib fragments typically twisted and used frequently for cauldrons, the sample of whose end was examined in its cross-section, too.

In the case of the chain link, the longer side of the fragmented piece selected for examination remained intact, it contained even a ring. Its size made it suitable for embedding the entire chain without sampling and examining the cross-section of bars via the slight destruction of the object. The examination spot seemed suitable for examination of both the production technique and characteristics of the composition.

Goal and sequence of examinations

Our examinations focussed on the determination of the composition of alloy used, which is a relevant piece of information for the description of the characteristics of the making. The elemental composition of samples was determined by means of SEM-EDS (**Figs. 6-7, 9-10.**). We made use of an optical microscope for the metallographical examination of the finds, to study the microstructure of the objects. Based on the features, description of the microstructure, we determined the technique and characteristics of making.

Fig. 6.: The microstructure of the cauldron-fragment. Recrystallized grains and intensive corrosion revealed by the optical micrograph. The corrosion can be discovered mainly at the grain boundaries. Zeiss Axio Scope.A1. Magnification.: 50x. Etching.: $FeCl_3$.,Bright Field

6. ábra: A bogrács töredék mikroszerkezete. Újrakristályosodtt szemcséket és intenzív korrózió nyomait látjuk a mikroszkópi felvételen. A korrózió nyomai leginkább a szemcsehatárok mentén fedezhetők fel. A felvétel Zeiss Axio Scope A1 mikroszkóppal készültek világos látótérben, 50x optikai nagyításban, $FeCl_3$ maratás után.

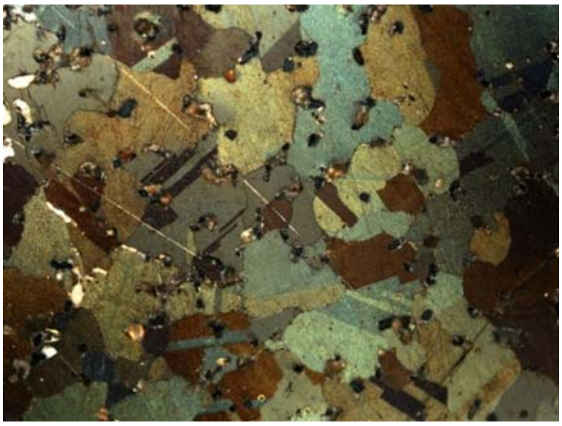

Fig. 7.: The grain structure of the cauldron in higher magnification. The colour etching reveals clearly the grains and the twin boundaries. The spots show the inclusions or corrosion products. Zeiss Axio Scope.A1. Magnification.: 100x. Etching.: K_2CrO_4.,Polarized.

7. ábra: A bogrács mikroszerkezete nagyobb nagyításban. A színes maratásban az iker határok jobban kivehetők. A fekete pontok zárványokat vagy korróziós terméket jelentenek. A felvétel Zeiss Axio Scope A1 mikroszkóppal készültek polarizált megvilágításban, 100x optikai nagyításban, K_2CrO_4 maratás után.

The samples taken out from the selected archaeological finds were embedded in a cold-curing two-component resin. The samples were ground using grades 220, 600, 1200 for flat surface specimen at 300 rpm, then polished using polishing tissue of 3 and 2 µ at 150 rpm. The samples prepared so were immersed in $FeCl_2$ and K_2CrO_4 etching agents for the analysis of grain structure. The goal of using two enchants was twofold. During etching with K_2CrO_4, an optically active oxide layer is formed on specific grains with a thickness varying depending on their crystallographic orientation. The grains obtain differing colours in polarised light. By way of this, one obtains outstanding information about the grain structure (whether it is a cast, formed, annealed grained structure). By examining in bright field, the sample prepared with potassium chromate, the existing inclusions can be well seen (e.g. copper sulphide inclusions) as well. The analysis of inclusions also provides important information on the making of the specific object or even its use. However, etching wit potassium chromate is sensitive to the combined etching of phases with different material grade. In such a case, etching with $FeCl_2$ makes grain structure optically sharper and more robust. Thus, it is less sensitive to enrichments and changes in composition caused by corrosion effects.

Samples examined recently

Fragment of the cauldron edge (Regszil1a)

Taking the average composition of the sample taken from the cauldron, it is a high tin-containing bronze. With consideration for the 11.3 w% of tin and 1.5 w% of nickel, a homogeneous solid solution phase is expected in the microstructure. As concerns its composition, this alloy has a high strength and hardness. It is still fit for cold-forming, with a high deformation resistance, though. For such an alloy, a high degree of plastic deformation, suggested by both the shape of the object and the hammer marks observed near the bottom of the cauldron excavated in 1907, cannot be imagined without annealing. The optical micrograph examinations revealed an annealed grain structure (**Fig. 6.**), just as did that of the cauldron found in the past. It is well visible in the micrographs taken in polarised light that the majority of sample is built by grains falling in the size range of 100 to 150 µm. In correspondence with earlier examinations, few but well distinguished twin-boundaries point out that the grain structure is annealed. A bent boundary surface can be observed in several places, which refers to the process of grain coarsening occurred due to increased temperature. The coarsening of the re-crystallised grains took place either during annealing or during use. This is a natural process but as was revealed by preparation using iron chloride, the grain size at the edges of the sample was well smaller (40 to 70 µm). Here, we need to consider two processes. The raw material is hard, difficult to form, thus, however thick is the formed part, its transformation needs significant energy or power. If the transformation is not successful, the material near the surfaces in contact with the tool or counterpart undergoes stronger deformation than the internal parts. This gradient already present in the deformatedness appears even during annealing, as tinier grains come to being near the surface then inside the sample volume. The degree of grain coarsening occurred in terms of the grain area does not depend on the original grain size but time and temperature. However, naturally due to constraints of geometry, one experiences other characteristic in the measured grain diameter. In addition, one can observe at the edge of sample in the microstructure revealed with iron chloride that the areas surrounding the surface are starkly corroded. Mostly corrosion along grain boundaries can be identified. The cauldron, in particular attention to its use, spent time in oxidising atmosphere at an elevated temperature. Oxygen tends to diffuse towards the internal volumes of the material along grain boundaries during the oxidation of copper alloys, and precipitate on the grain boundary as tiny phases of Cu_2O. If the object is subject to later corrosion effects, this phenomenon is able to promote corrosion along grain boundaries. Moreover, it can slow down grain coarsening process near the surface. All in all, it can be determined considering the circumstances of the excavation mentioned in the introduction to the finds, that both sides were subject to corrosion effects in the soil in most of the long period spent there. It can be well seen, however, that there is a noticeable difference for the degree of corrosion between the two sides. With a view to the circumstances of the excavation, the entire object can be regarded as to have been exposed to the identical corrosion effect, therefore the reason for the difference can be sought in the "history" of the sample. In the light of the composition of the material and the concluded properties, this can be the result of, on the one hand, the two different deformations on the two sides of the sheet. Considering the geometry of the object, this assumption can be backed by the fact that the shape and design carries this difference intrinsically. This is complemented by the technique of the period and the hardness of the metal. On the other hand, it is the use of the object. One side of the cauldron is in direct contact with the fire, so it is exposed to higher heat there. Depending on the fire, the oxidising effect can also be different, but we draw here attention to the fact that we examine the flange where these differences are by all means of lower degree than in the case of the cauldron body. However, if we take this into account, we can expect a corrosion along boundaries with a different rate of corrosion due to the differing corrosion effect. In our present study, we see a more powerful effect in the forming technology based on our examinations. Our data indicate that the sheet was subject to an asymmetric formation (the formation was stronger from one side) and the transformation of the sheet had not taken place during the plastic deformation prior to the last annealing. Therefore, smaller-sized grains developed along this surface during the re-crystallisation taking place in the annealing. The effects occurring during the mentioned oxidation, which slow down coarsening, could have intensified this difference during use. The differing rate of corrosion of the two sides could have been caused jointly by this difference and the two sides subjected to uneven oxidising effect. As mentioned, this coincides to what we may conclude from the shape and normal use of the object.

As was already mentioned, the analysis of the inclusion bears great importance, too. The presence of homogeneous solid solution can be expected, however, foreign phases can enter the metal both during smelting and making that can tell about the way of manufacture. As was shown by preliminary examinations, copper sulphide inclusions were found in the grain structure in the optical micrographs (**Fig. 7**.).Their size is about 20 µm, their colour being dark grey on the optical micrograph. Their shape typically have a rounded boundary, mostly of globular nature. This make us conclude that the material spent an extended time at an elevated temperature after smelting. With consideration to the use of the object, we cannot draw any conclusion as to what technique was used for the making. Literature sources and earlier examinations point out that their origin derives back to smelting. SEM-EDS analyses revealed that the inclusions have measurable sulphur content in addition to copper, but no tin can be detected. Tin can be measured in metallic areas only. No nickel content can be detected even in some of the sulphide inclusions. In specific inclusions nickel can be detected but we note that copper is able to dissolve nickel in unlimited amounts, which is an important factor in this case. Phases with high tin and high nickel content can be mostly located in the proximity of sulphide phases (**Fig. 8.**).

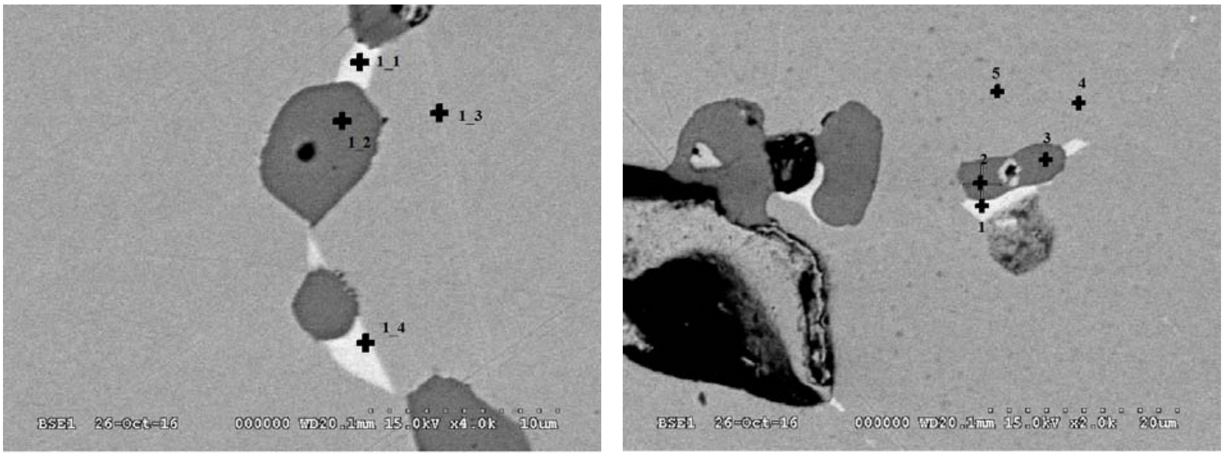

Fig. 8.: SEM micrographs taken from the cauldron. The micrograph shows the inclusions. The large phases are copper-sulphide inclusions, the small bright ones are SN-Ni rich phases.

8. ábra: A bogrács töredék éléből vett minta SEM felvételei. A felvételen a zárványok láthatók: a nagyméretű fázisok réz-szulfidok, a kisméretű világos fázisok az Sn-Ni gazdag fázisok.

Table 1.: Results of the local EDS analysis of cauldron edge

1. táblázat: A bográcstöredékből vett minta helyi SEM elemzéseinek eredményei

	Atomic percent (%)					
	C	O	S	Ni	Cu	Sn
Fig8a: 1_1				9.43	69.19	21.38
Fig8a: 1_2		1.93	30.19		67.89	
Fig8a: 1_3		3.57			90.66	5.77
Fig8a: 1_4				2.38	90.50	7.13
Fig8b: 1	13.53	10.75		8.57	48.55	18.60
Fig8b: 2	7.86	2.81	24.82	0.75	63.76	
Fig8b: 3	9.09	41.59		0.18	35.73	13.41
Fig8b: 4	8.90	6.82			79.36	4.92
Fig8b: 5	10.09	3.32		1.27	80.33	4.99

The SEM-EDS study provides only compositional data, whose evaluation requires the consideration that the examined phases are smaller than the excited volume, thus, the composition of the solid solution in the surrounding of the phase distorts its composition. However, it appears from the data that nickel content is about 9 w% and tin content is not lower than 18 w% (**Table 1.**). Copper dissolves tin both in liquid and solid state very well and dissolves nickel in unlimited amounts. (ASM Handbook 1994, 318) Based on thermodynamic relations, a phase can remain in this state only if it is an intermetallic compound with a high melting temperature of the tin nickel alloy system. These are Ni_3Sn_2 and Ni_3Sn_4. Although it cannot be supported by the presented examinations and is only a conclusion but it can be either of them based on the compositions, but Ni_3Sn_2 seems to be more probable, supported by its melting temperature. However, if copper and tin had been smelted together, the development probability of this compound based on the above dissolution tendencies is insignificant as the professional literature does not refer to this phase either. Yet, it develops easily in the tin nickel system. This leads to the assumption that metallic tin was alloyed into copper in this case. A part of the nickel content had been introduced in the alloy along with tin just as had been the mentioned compounds. Their tinyness is explained by the fact that these compounds dissolve slowly in copper and their tin and nickel content is transferred into a solid solution. However, the alloying and afterwards the crystallisation period had not been long enough to get dissolved all, the examined sample shows the remainder. This is backed also by the fact that the largest part is located near the copper sulphide. Copper sulphide is a phase with low melting temperature and solidifies in the last stage of crystallisation, thus, the inclusions with high melting temperature not able to dissolve in the melt shall be located here. These observations all point out that direct alloying had been applied for the making the material of the cauldron, the metal-state copper was molten together with metallic tin.

Fragment of the incenser chain link (Regszil_1)

Optical micrographs revealed a dendritic structure. This refers to the making of chain link by casting. Based on the large size of the dendrites and their radial arrangements in the cross-section, the melt had not been overheated much over its melting temperature. The fine structure of dendrite arms refers to high cooling rate, which arises from the dimensions of the object.

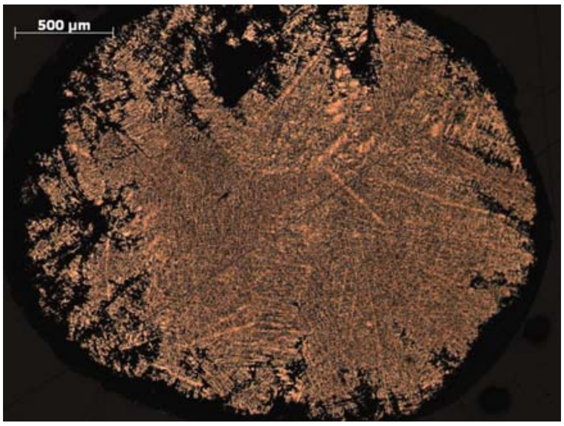

Fig. 9.: The microstructure of a chain fragment. Dendrites with fine secondary arms can be seen on the micrographs. Zeiss Axio Scope.A1. Magnification.: 50x. Etching: $FeCl_3$, Bright Field

9. ábra: A lánctöredék mikroszerkezete. Kis szekunder ágtávolságú dendrites szerkezet látható a felvételen. A felvétel Zeiss Axio Scope A1 mikroszkóppal készültek világos látótérben, 50x optikai nagyításban, $FeCl_3$ maratás után.

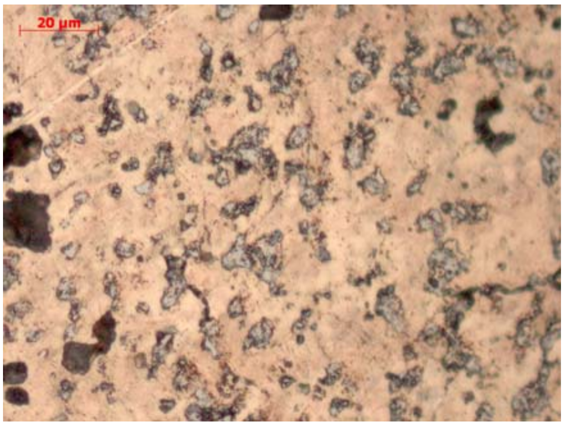

Fig. 10.: The microstructure of a chain fragment in higher magnification. A lot of inclusions can be seen on the micrographs. Zeiss Axio Scope.A1. Magnification.: 1000x. Etching: $FeCl_3$., Bright Field.

10. ábra: A lánctöredék mikroszerkezete nagy nagyításban. A mikroszerkezetben sok zárvány fedezhető fel. A felvétel Zeiss Axio Scope A1 mikroszkóppal készültek világos látótérben, 50x optikai nagyításban, $FeCl_3$ maratás után.

The raw material is bronze, which, in addition to its tin content above 5% contains a considerable amount of lead (14%). The addition of large amount of lead had been necessary for the sake of the high ability of the thin chain link to fill in the mould (**Fig. 9-10.**).

Electron microscopy reveals significantly heterogeneous structure where globular white phases are the drops of the metallic lead. Several studies highlight that copper dissolves lead neither in liquid nor solid state, thus, it creates separate globular phases. In addition, one can encounter here lower quantities of copper sulphide inclusions from smelting.

The material of the chain link similarly includes high tin- and nickel-containing phases just as in the case of the cauldron (**Fig. 11.**). The tin nickel ratio also in this case assumes the presence of the mentioned tin nickel intermetallic phases (**Table 2.**). Also here, these tiny phases can be found in small quantities, which supports that low nickel is present in the system and it had been incorporated in the raw material of the chain link as the contaminant of the lead during alloying.

The examination of the chain link exhibited the same material-specific features as also in the case of the object made by the other basic technique for making (casting) as was seen in the case of the object with larger weight formed by hammering, the cauldron. At the same time, the addition of lead is specific to the production technique. This is all supported by the fact that the presence of tin nickel intermetallic phases is the feature of the raw material for general use, the specialty of the bronze raw material. Also, this points out the metal-to-metal alloying.

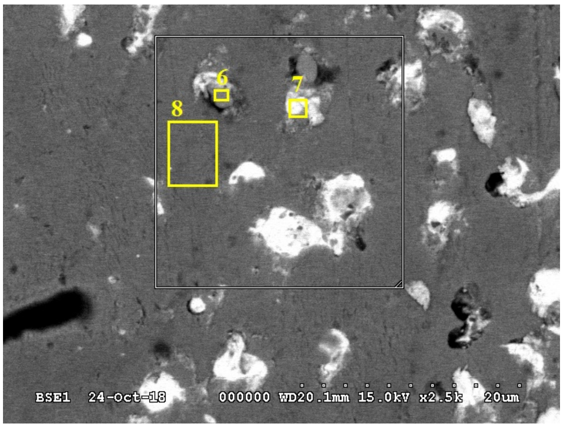

Fig. 11.: SEM analysis of the chain fragment reveals that the majority of the inclusions are lead drops (white phases)

11. ábra: A lánctöredék SEM vizsgálata rámutatott, hogy a zárványok jelentős része ólom csepp (fehér színű fázis)

Table 2.: Results of EDS analysis of different phases in chain link

2. táblázat: A lánctöredék különböző fázisainak EDS elemzése

Chemical composition. wt %						
	C	O	Ni	Cu	Sn	Pb
Spectrum 6	4.10	2.41	7.24	50.96	25.26	10.03
Spectrum 7	12.03	5.55		10.44		71.98
Spectrum 8	1.14	1.73		86.35	10.78	

Evaluation of the results of metallographical examinations

We carried out metal analyses in the case of the cauldron found in Regöly, on the material of the cauldron, the rivet holding the nib and the nib itself. The goal of the analyses was to determine the raw material and the way of making. Therefore, we performed optical microscopy (OM) and electron microscopy (SEM) analyses on the flat polish specimens of the samples taken from the objects. It was determined that the cauldron was made of a high lead-containing raw material, which had presumably been required to increase strength and this is supported by the wall thickness of the cauldron. The cauldron had been formed by hammering and made by an intermediate annealing. This is shown by both the dimensions and design of the cauldron. Due to high tin content, the raw material is hard and still can be cold-formed though, but the large deformation necessary for achieving the wall thickness of the cauldron by applying annealing, whose traces are reflected in the microstructure as well. The analysis of the corrosion effects highlighted that one side of the cauldron had undergone a stronger deformation-formation. This is also supported by the shape of the cauldron as the base plate had to be deepened. We measured a significant quantity of nickel in the raw material composition besides tin. This should not be a surprise based on parallelisms, but we encountered tiny tin nickel inclusions in addition to the copper sulphide inclusions usual of origin of smelting. Considering the thermodynamic behaviour of the alloy, this phase can be found only if the tin nickel is an intermetallic compound with high melting temperature. This is backed by its measured composition, too. This intermetallic compound can, however, be included in the copper raw material by direct alloying during adding lead if the intermetallic phases had been developed in the tin prior to alloying. This clearly indicates direct alloying of the raw material of the cauldron and requires re-thinking about the source for the ores used. The high nickel content provides a safe fingerprint for the search of the tin source (**Fig 8., 11., Tables 1-2.**).

Archaeological parallels, historical background of the cauldrons of Regöly

The research grouped cauldrons, similar in form to the ones from Regöly, as type C for decades, or created local groups based on that one. (Merhart 1952, 61; Egg &Kramer 2013, 243-255, Abb. 104; Patay 1990, 25–30; Jereb 2016, 105). Today it is clear however, that the traditional grouping method is not sensible for cauldrons produced in serial manufacturing, sometimes even made from different parts. Studies had been written on the subject earlier, presenting the actual significant parallelism (Szabó 2009, 348; 2012, 86; 2013, 296; Fekete & Szabó 2017a). Therefore, these categorizations are not considered in these examinations with a technological aspect. The passage from Bronze Age to Iron Age refers to a historical, ethnical changeover underlying in the background of the serial production appearing in the second half of the 7th century BC in the Carpathian Basin that these products and the histories of technologies used for their production show orientation towards Asia Minor. Thanks to those arriving from there in several waves, Europe had been invaded by an elite belonging in part relative nations/cultural medium and their armed escorts via the Carpathians, Balkans, Italy and the Western Mediterranean Basin (Horváth & Szabó 2017). At the time of the first Eastern waves, the Urnfield culture and the early Hallstatt culture, a dispersed appearance of the new object types is typical (Fekete & Szabó 2015; Szabó 2017b). In the light of newer direct Regöly finds showing relations with Asia Minor, not to be explained by commercial relations, it can be well seen that metal workmanship based on workshop practices and technological know-how of the Ancient East had been radically transforming the local sheet industry of Bronze Age origin from the second half of the 7th century BC and it had been already producing its products locally as well. At this time, it not only concerned copying object shapes to be observed in the Urnfield period but an exact knowledge and application of new technological processes and work organisation. This indicates that those arriving in this age in recurrent waves via various paths and with different destinations had brought their craftsmen as well, as it was shown in other fields by technological changes occurring explosively at almost at the same time across Europe such as in iron making or pottery. One important element of these changes was the phenomenon observed first on Regöly cauldron and chain link fragment that they had already begun to alloy copper with metallic tin during bronze making. According to Lloyd Weeks, the metallurgy and commerce of tin is one of the most disputed issues of the manufacture of bronze (Weeks 2004; 2012, 303). After some smaller objects appearing from the middle of 3rd millennium BC, the commerce of metallic tin appears only in the second half of the 2nd century BC in Assyrian texts. At this time, an already considerable amount could have been in circulation as was shown by the ship of Uluburun sunken around 1300 BC where over a ton of metallic tin ingots was found (Weeks 2012, 303-305). It appeared in minor quantities in Europe at around this time as was indicated by the Swiss finds (Primas 1984). Mária Fekete drew the attention to its application in the early Iron Age of the Carpathian Basin, the ornamentation of clay vessels with tin foil (Fekete 1981, 151-152; 1985a, 61-63; 2007, 61). However, we had no data for the use in its metallic state for alloying similarly pure metallic copper in Europe so far.

The way copper is alloyed in the HaD$_1$ period in the case of examined cauldrons and chain links

According to our measurements, the difference between the cast sling of cauldron excavated from Regöly, Hallstatt burial site no. 696 and Býčí skála Cave is lower than 5% at each point, which indicates the use of identical matrice or mould (Szabó 2013, Pl. 6. 1). The serial production is just as a relevant issue in the finds material as the separation of products made in the same workshop provides an opportunity to shed light more exactly on relations in history and time-scale. The use of bronze casting, matrices and moulds itself carries intrinsically the opportunity of the serial production from earliest times and the craftsmen of the period made use of this for smaller serial production mainly for local use. From the early Iron Age, however, in part parallel to the Greek colonisation, extremely similar objects appear across Europe, among which often only just some small parts show a great extent of similarity (Kimmig 1983). Exactly in the era of changeover from bronze to iron, a new phenomenon can be encountered: certain objects are assembled from components produced in serial production. This is already a higher level of workshop activities and shows towards the manufacturisation, whose origins can be found in the Ancient East and may refer in its background to the societal structure different from the state of Europe at the end of Bronze Age. The data referring to the direct alloying of copper unknown earlier in the European material of the current archaeometallurgical examination of the cauldron fragment of Regöly and the serially produced censer chain link based on a similar work organisation also proves the earlier observations (Fekete & Szabó 2015, 2017a; Kürthy et al 2016; 2018), that manufacturisation and the accompanying explosion-like technological advance are not the outcome of the internal development of the Hallstatt culture sphere. It had been a ready-implemented practice in Europe by masters of the last Ionian migration starting from Asia Minor in the last third of the 7th century BC into the Carpathian Basin just as had been the technology of alloying copper directly with metallic tin, one of whose main proofs is the Sn-Ni phase observed on our objects recently in their raw material.

Table 3.: Chemical analysis and identification of the Ni-Sn phases by its composition and the data from the equilibrium phase diagram

3. táblázat: A Ni-Sn intermetallikus fázisok kémiai elemzése és azonosítása az egyensúlyi fázisdiagram összetételi adatai alapján

measure	Sn (solid solution)	Ni	Sn	Sn (phase)	Sn/Ni	phase
weight %	11.3	8.66	37.44	26.14	1.49	Ni3Sn2 (1.35)
weight %	11.3	9.56	37.47	26.17	1.35	Ni3Sn2 (1.35)
atomic %	5.77	9.43	21.38	15.61	1.66	Ni3Sn4 (1.33)
atomic %	4.92	8.57	18.6	13.68	1.60	Ni3Sn4 (1.33)
atomic %	4.99	2.33	7.13	2.14	0.92	Ni2Sn2 (0.67)

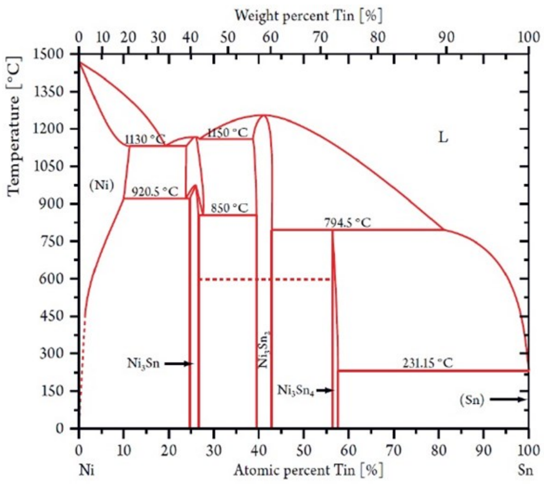

Fig. 12.: Sn-Ni Phase Diagram

12. ábra: Az Sn-Ni egyensúlyi fázisdiagram

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