# PREPARATION OF RAPIDLY QUENCHED BILAYER RIBBONS, THEIR PROPERTIES AND INTERFACE STRUCTURE


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## 1. Introduction

Rapidly quenched metallic materials are attractive for their physical properties related to amorphous or subsequently (nano)crystallized structure. They represent a kind of material with promising applications potential. Planar flow casting is a widely used technique of their preparation in form of long ribbons several millimeters or tens of millimeters wide and about 20-30 μm thick. Demand for bulk amorphous materials stimulates the search for preparation of thicker ribbons in the form from bilayers to multilayers. This approach opens in spite of relatively limited material thickness a new challenge of possibility to prepare a heterostructures by changing chemical composition of the individual layers. Such materials, especially in the case of bilayers from different alloys, are interesting for their intrinsically graded properties, which can lead to specific applications, or objects for investigation of various phenomena and processes at material interfaces.

In our work a previously reported rather complicated technique of melting two different alloys separately in two crucibles [1] has been replaced by the technique of a single crucible with two nozzles close to each other and with a partition between them forming two separate vessels [2,3]. Such an arrangement allows easy formation of two homogeneous layers along the whole ribbon length. The aim of the work has been to investigate the formation and character of the bilayers, the connection of the two layers and the mutual intermixing of components at the interface between the layers. Magnetic properties of bilayers have been checked also with respect to potential applications of such materials.

## 2. Experimental

Rapidly quenched bilayer ribbon with high quality surface and edges consisting of $Fe_{77.5}Si_{7.5}B_{15}$ (air side of the ribbon) and $Co_{72.5}Si_{12.5}B_{15}$ (wheel side) layers has been prepared by planar flow casting from a single crucible using the double-nozzle technique. Ribbons with typical thickness of 45–50 μm (~ twice the thickness of the usual single-layer ribbons) and 6 mm width exhibited amorphous structure of both layers according to X-ray diffraction (Cu Kα radiation) in as-quenched state [4], identical to that of the single-layer ribbons with the respective compositions, which have been prepared as well via single-nozzle casting. Temperature dependences of electrical resistivity R(T), and "magnetic" weight (thermogravimetry, TGA, with weak permanent magnet) have been measured in amorphous state and during crystallization of both layers, which take place at different temperatures (Fig. 1.). The structure of the interlayer has been investigated by cross-sectional transmission electron microscopy (CS-TEM), in scanning mode (STEM) coupled to analysis by high angle angular dark field (HAADF) detector, energy-dispersive X-ray spectroscopy (EDX) and electron energy loss spectroscopy (EELS). Samples for CS-TEM have been prepared by high-precision mechanical polishing followed by ion beam milling.

## 3. Results and discussion

Temperature dependence of electrical resistivity of bilayer ribbons until ~800K (while both layers are still in amorphous state) exhibits the usual character of metallic glasses – high specific electrical resistivity of ~130±10 μΩcm (similar in either of the layers, as determined from measurements on single layer ribbons) and low temperature coefficient of electrical resistivity. Crystallization starts above 800K in the Fe-Si-B as witnessed by the decrease of electrical resistivity (corroborated by the R(T) of single-layer ribbons in Fig. 1a); the Co-Si-B layer starts to crystallize above ~850K. The remaining amorphous matrix in both layers transforms in the vicinity of 900K. TGA signal in weak magnetic field exhibits a decrease to nearly zero at ~720K due to the Curie temperature of the amorphous Fe-Si-B (Curie temperature of amorphous Co-Si-B is ~450K, as shown in Fig. 1b). The increase of the magnetic TGA signal above 800K correlates well with the formation of soft magnetic crystals of bcc-Fe(Si). Its further increase above 850K reflects the formation of Co-rich phases in the Co-Si-B layer.

We have used the relative proportions of the measured inverse electrical resistivities of single layer Fe-Si-B and Co-Si-B ribbons with cross-sections 0.48/0.52, respectively, determined by compositional line analysis, to compute the inverse electrical resistivity of the ideal parallel combination of the two layers. The calculated the R(T) dependence shown in Fig. 1a) is nearly identical to that of the original bilayer. This indicates that the two constituent layers of the bilayer really have a nearly identical thickness as well as that there is only a minimal deviation from the superposition principle for electrical conductivities of parallel conductors. It suggests that the intermixing in the interface region is on very low level and does not change significantly even after complete crystallization of the layers upon linear heating. Similar effects and behaviour have been observed by measurements of magnetic TGA.

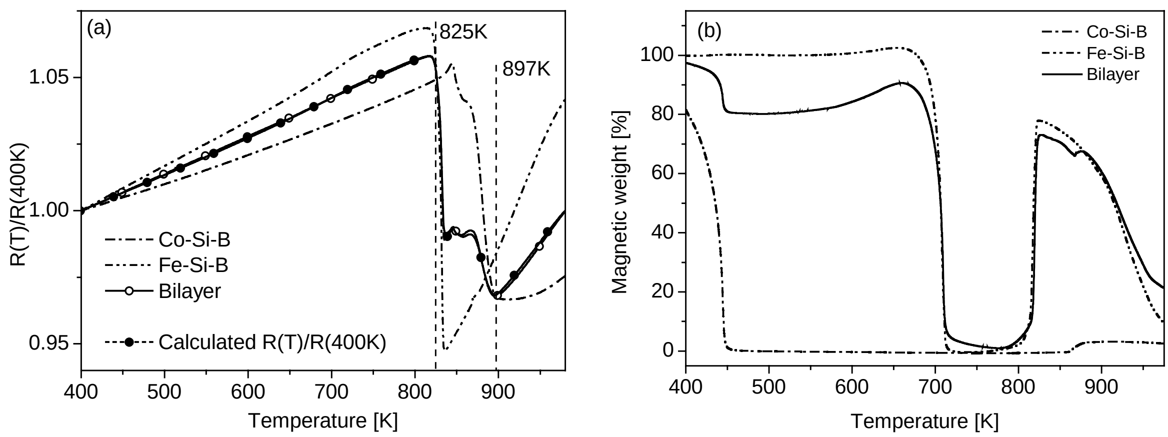


Fig. 1: *Temperature dependencies of (a) relative electrical resistivities of the single layer Fe-Si-B and Co-Si-B samples and the calculated total electrical resistivity of the parallel combination of the two single layer samples compared to that of the measured bilayer ribbon;(b) magnetic TGA signal of the single layer Fe-Si-B and Co-Si-B samples and of the bilayer sample. The vertical lines in Fig. 1a) indicate positions selected for detailed structural analysis of the interface. Heating rate was 10K/min.*

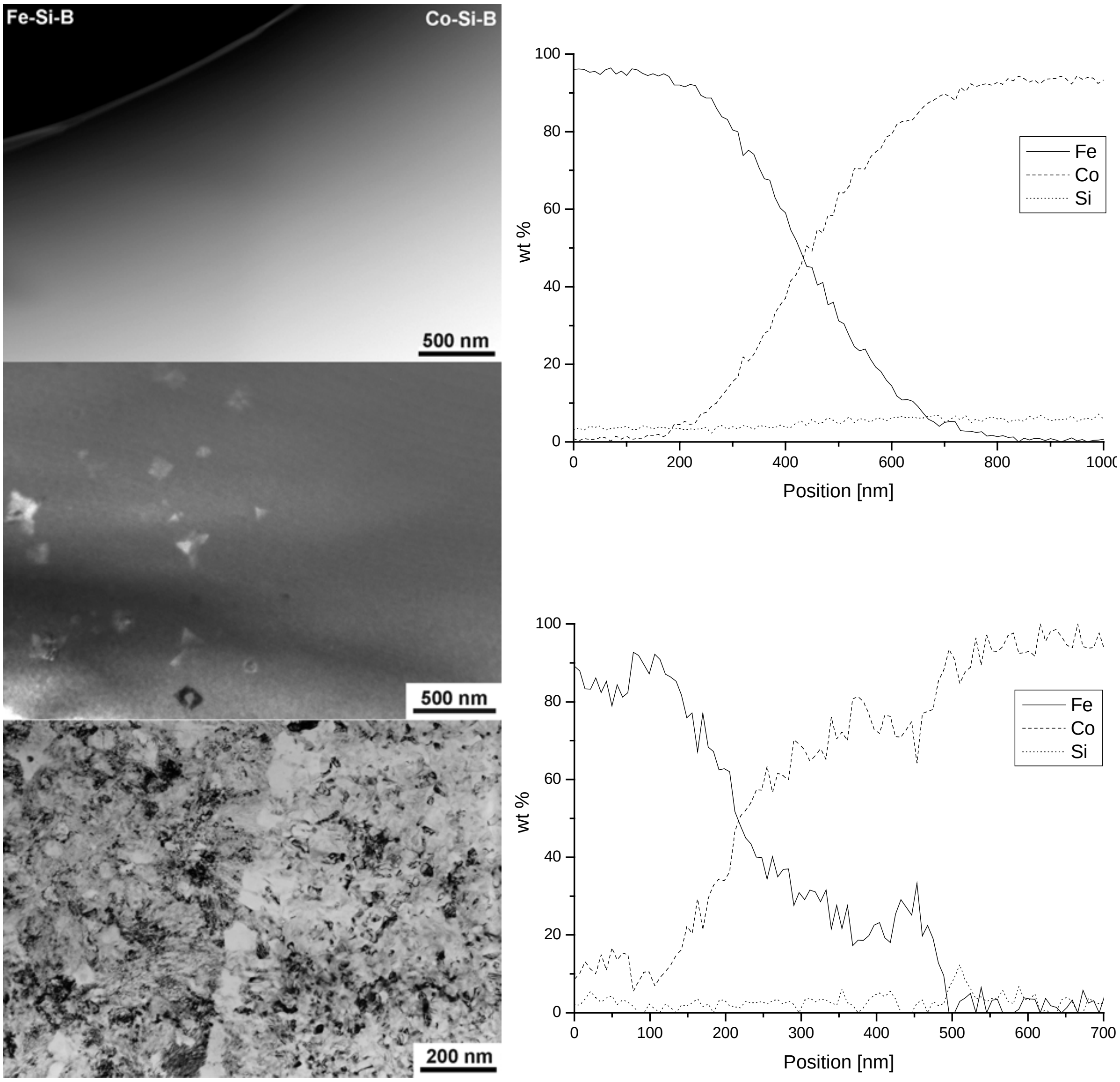


Fig.2: *CS-TEM analysis of bilayer interface:(left-up) HAADF image of as-cast, (right-up) concentration profile of as-cast , (left-middle) bright field TEM after linear heating to 825K, (left-bottom) bright field TEM after linear heating to 897K, (right-bottom) concentration profile after linear heating to 897K.*

CS-TEM observations of the as-cast bilayer sample in bright-field mode confirm its amorphous structure and shows contrastless interface between the constituent layers. HAADF analysis of interface allows observing of its planar and well-defined character (Fig.2 left-up). It has to be noted that the interface is represented by slight but well detectable drop of intensity in the middle of image. Corresponding concentration profile across the interface determined by STEM/EDX (Fig.2 right-up) gives the thickness of ~500nm. The STEM/EELS intensity profiles for constituent elements (not shown) yield the same value. Here it has to be noted that in order to eliminate the TEM sample (thin foil) thickness effect, the profile has been measured under the angle of ~35 deg. to interface normal, so its real thickness should be ~20% lower. For the sample after linear heating to 825K, which corresponds to the temperature after the onset of crystallization of Fe layer, the bright-field TEM analysis (Fig.2 left-middle) shows the separation between the entirely amorphous structure of Co based layer and partially crystallized structure of Fe based layer. A sharp interface has been observed also on the sample annealed to 897K (corresponding to fully crystallized bilayer structure - Fig.2 left-bottom), separating the regions of growing crystals of different morphologies for individual layers. The morphologies of crystallized layers correspond well to those observed during the analogical analysis of crystallization of single amorphous layers of the corresponding stochiometries [5]. Corresponding concentration profile across the interface determined by STEM/EDX (Fig. right-bottom) gives the similar value of interface thickness (~500nm), but its step-like character indicate a formation of intermetallic crystals in the interface region. However their structure still needs to be analysed.

## 4. Conclusions

Rapidly quenched bilayers in fully amorphous state in form of regularly shaped long ribbons with smooth edges were successfully prepared by rapid quenching using double-nozzle technique. From the results it seems evident that the process of connection of the two layers during preparation takes place by solidification with only a small extent of mutual interdiffusion of component atoms localized to a narrow well-defined and nearly planar interface, leading to mechanically solid connection between the two layers. Detailed CS-TEM analysis of the interface between constituent layers shows ist planar and well-defined character. The determined thickness of ~500nm is not significantly affected by bilayer annealing. A formation of intermetallic crystals in the interface region is indicated.

Bilayers with different composition have exhibited good conformity with the superposition principle for selected physical properties, namely electrical conductivity, in the whole temperature range investigated.

The character of the interface predestines the studied bilayers as suitable objects for further studies of the phenomena at the interfaces between two amorphous systems or between amorphous and crystalline phases. At the same time advantageous presence of two different magnetic materials in a compact bilayer allows their convenient application as magnetic sensors or actuators.

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