A New Method to Measure Temperatures in Polymer Melts Using Heusler Alloys*

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Synopsis

With the help of ferromagnetic Heusler alloys, which are mixed in powdery form into a cylindrical polymer sample surrounded by an induction coil, temperature can be measured. Some 10 K below the Curie temperature small temperature changes already yield considerable signals in the induction coil via the temperature dependence of the permeability of the alloy. From the tremendous variety of Heusler alloys available for each temperature region of interest the proper material can be chosen. The amount of sample needed is small enough to leave the rheological properties of the melt unchanged. Measurements do not involve touching the sample and follow each temperature change more or less simultaneously since the grains of the alloys are very small and have very small heat capacities only.

INTRODUCTION

Temperature measurement in a polymer melt is a formidable task especially using thermocouples. To measure temperature changes as fast as possible, the sensors have to be small (low heat capacity). As a consequence, they can be torn apart by melts of high viscosity because of their mechanical weakness. If they are mechanically strong, however, their response suffers from a considerable delay.

It was worthwhile therefore to look for new methods which would allow the measurement of fast temperature changes immediately and also avoid heat conduction through the wire of thermocouples which occurs by touching the sample. This can be achieved, in principle, by filling an induction coil with a substance whose permeability changes with temperature. A proper measuring cell based on the mentioned principle has been developed recently.[1] The new cell is part of a new magnetoviscosimeter.[2-4] More important than the cell, however, is the substance which can be mixed with a polymer melt (without a chemical reaction) and whose magnetic properties (permeability) change is sufficient enough in the temperature range of interest. In the case of iron it would be ca. 1000 K, much too high for polymer melts.

One of the authors had to do quite a lot with Heusler alloys in former times.[5,6] This group of mostly ferromagnetic alloys has Curie temperatures T_c's from 93 to 1150 K, depending on composition. For each temperature region a special Heusler alloy can be selected (see Table I). Near the Curie temperature (where ferromagnetism disappears) the susceptibility begins to fall rapidly changing the inductivity of a coil wrapped around the sample.

*Dedicated to Prof. H. Janeschitz-Kriegl on the occasion of his 65th birthday.

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TABLE I
Curie Temperature (K) of Heusler Alloys with Manganese

With manganese		Without manganese	
Co_2MnSi	985	Co_2FeGa	1150
Co_2MnGe	905	Fe_2CoGa	1050
Co_2MnSn	829	Ru_2FeSn	593
Co_2MnGa	694	Co_2ZrSn	444
Cu_2MnAl	613	Co_2HfSn	394
Cu_2MnSn	530	Co_2TiSn	359
Cu_2MnIn	520	Co_2VGa	349
Rh_2MnGe	450	Co_2VAl	310
Rh_2MnSn	410	Co_2HfAl	193
Ni_2MnGa	379	Co_2HfGa	186
Ni_2MnSb	360	Co_2ZrAl	185
Ni_2MnSn	344	Co_2TiAl	138
Rh_2MnPb	338	Co_2TiGa	130
Ni_2MnIn	323	Co_2NbSn	119
Pd_2MnSb	247	Rh_2NiSn	93
Au_2MnAl	200		
Pd_2MnSn	189		
Pd_2MnGe	170		

Using powdery alloys (particles as small as 1 μm) a temperature change of the melt (with the powder) is converted into a signal on the induction coil immediately.

EXPERIMENTAL

As can be seen from Table II already, there are many elements from which the various Heusler alloys can be synthesized. By changing the composition of the alloys for each Curie point a tailored material can be prepared. As an example, the system $(Ni_xCo_{1-x})_2MnSn$ was taken; by replacing Ni with Co different T_c's can be achieved. The magnetic properties of these substances were described by one of the authors.[6] In this system there exists a linear relationship between T_c and the composition (Fig. 1). Furthermore, the temperature range of T_c covers a region which is very interesting for a variety of industrially applied polymers.

TABLE II
Preferred Positions of Different Elements in Heusler Alloys with the Composition X_2YZ

	Y					*X*					*Z*			
	IIIB	IVB	VB	VIB	VIIB	VIIIB			IB	IIB	IIIA	IVA	VA	VIA
1														
2											B	C	N	O
3											Al	Si	P	S
4	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se
5	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te
6	La	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po

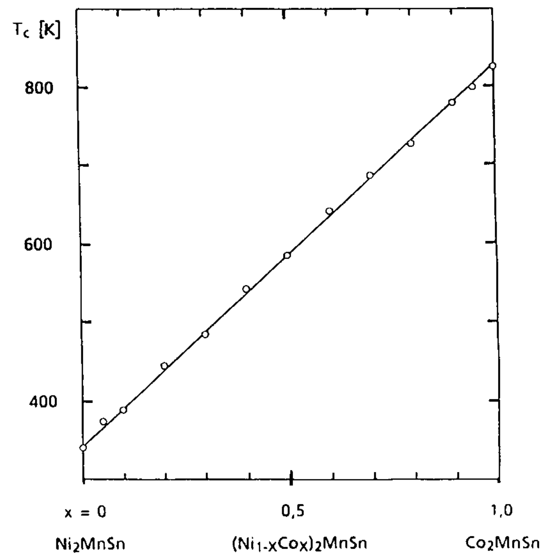

Fig. 1. Curie temperatures of the system $(Ni_{1-x}Co_x)MnSn$ as a function of composition, expressed by the parameter x.

Another advantage is the cheapness of the components used in the system and the comparatively simple preparation method as demonstrated by a characteristic example.

The proper amounts of powdery components are weighed, mixed, and pressed to pills. These are sealed in evacuated silicon tubes. Since tin has a melting point (232°C), which is much below the melting points of the other components (ca. 1000°C), a special temperature program has to be applied (6 h at 200°C, 12 h at 230°C, and 12 h at 300°C). After the tin has reacted with the other components, the sample will be annealed at 800°C for 3 days.

The proper measuring coil wrapped around the cylindrical sample is part of an electric circuit in which the inductivity is compared with the one of a reference coil. Temperature changes below T_c yield a differential signal which is amplified. A more extended description of the whole circuit can be found elsewhere.[1]

RESULTS AND DISCUSSION

In Figure 2 reduced signals for different compositions are shown. One mixed phase covers a temperature region of about 10–20°C, in which its behavior can be approximated by a linear function. To cover a more extended region, two or more phases have to be mixed in proper proportions. To get the results of Figure 3, this proportion is 1 : 1 : 1. By choosing carefully different phases with other T_c values and varying proportions, a linear relationship between height of signal and temperature can be obtained. In this example a signal of 90 mV was achieved with 100 mg alloy, respectively 0.1 mV/mg °C. Taking into consideration the volume of the sample within the measuring coil, 10 vol. % was needed for said signal. Besides temperature measurements in polymer melts it could be very interesting to follow the exothermic reaction during the buildup of the network of epoxy resins.

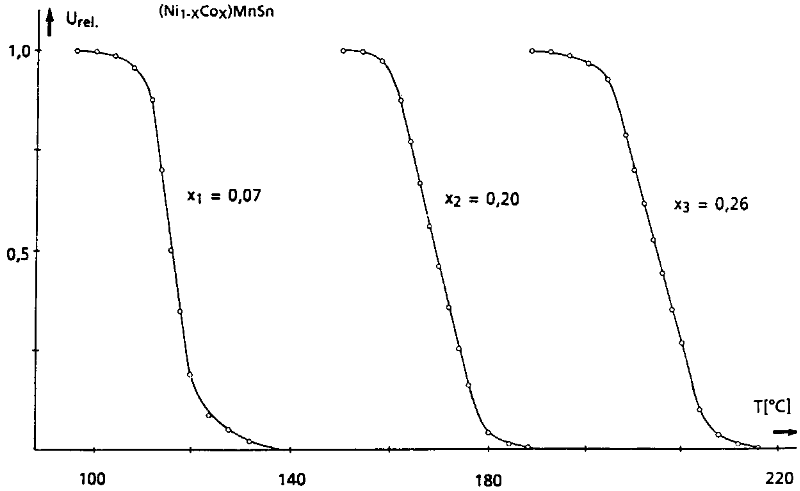

Fig. 2. Reduced signals U_{rel} as a function of temperature for three different compositions x of the system ($Ni_{1-x}Co_x$)MnSn ($x_1 = 0.07$, $x_2 = 0.20$, $x_3 = 0.26$).

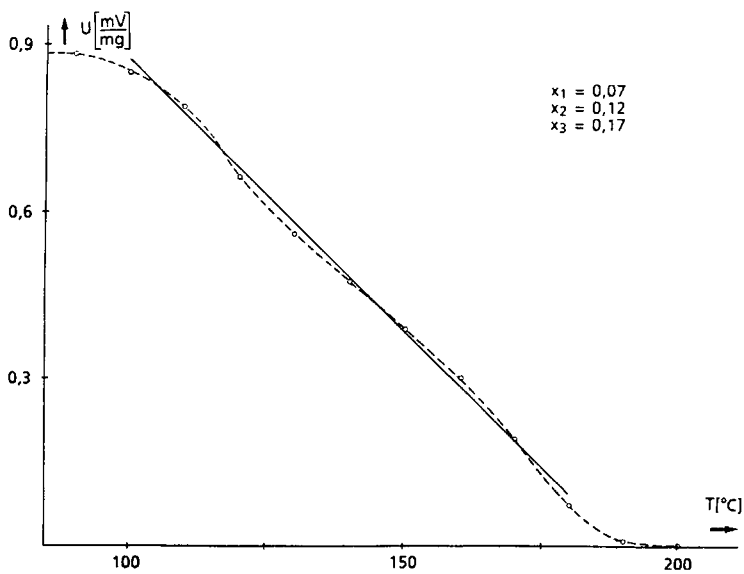

Fig. 3. Temperature dependence of the signal produced by a 1:1:1 mixture of three different compositions x_1, x_2, and x_3 of ($Ni_{1-x}Co_x$)MnSn ($x_1 = 0.07$, $x_2 = 0.12$, $x_3 = 0.17$).

Thermocouples cannot be removed after hardening of the sample whereas specimens with the cheap powdery Heusler alloy as a temperature probe can be thrown away simply after the experiment.

Further applications:

a. To get an idea about the temperature distribution along an extended sample several coils can be wrapped around and switched on and off to feed the differential amplifier one after another.

b. Viscous fluids which do not mix very fast are suited for the measurement of radial temperature distributions by mixing the powder into certain layers only (which can be changed, of course).

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References

1. W. Hermann and R. Sobczak, *J. Appl. Polym. Sci.*, **37**, 2675 (1989).
2. R. Sobczak, *Rheol. Acta*, **25**, 75 (1986).
3. W. Hermann and R. Sobczak, *Mh. Chem.*, **117**, 753 (1986).
4. M. Gahleitner and R. Sobczak, *Rheol. Acta*, **26**, 371 (1987).
5. R. Sobczak, *Mh. Chem.*, **107**, 977 (1976).
6. R. Sobczak, *Mh. Chem.*, **109**, 453 (1978).
7. R. Sobczak, *Mh. Chem.*, **109**, 1423 (1978).

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