Hysteresis in Magnetocaloric Materials: An experimental and modelling approach

In this thesis the effects of hysteresis on magnetocaloric material properties and their performance in magnetic refrigeration devices are investigated. This is done through an experimental and model study of first order magnetocaloric materials MnFe(P,As) and Gd5Si2Ge2. The experimental characterization of the magnetocaloric effect (MCE) in these materials is done through conventional indirect magnetometric and calorimetric methods, as well as newly developed direct methods. The determination of the MCE due to a magnetic field change is in principle given by the isofield material entropy curves, obtained at the initial low and final high field. However, in first order materials thermal entropy hysteresis loops are obtained through characterization, corresponding to measurements done in an increasing and a decreasing temperature mode. Indirectly determining the MCE through the use of the Maxwell relation or calorimetric measurements done only in a heating or cooling mode, estimate the MCE as the reversible difference between the set isofield heating-heating or cooling-cooling entropy curves. Here it is shown that direct measurements suggest that the real MCE is given by the difference between the low field heating and high field cooling entropy curves, which can reduce the MCE estimate significantly. The experimental data obtained through the material characterization is used as a foundation for Preisach type models. This type of model is suited to handle the non-equilibrium nature of first order materials, taking the magnetic and thermal history dependence of material properties into account, as well as the heat production due to hysteretic losses. MnFe(P,As) and Gd5Si2Ge2 compounds are modelled and it is found that the Preisach approach is suitable to reproduce material behavior in both cases. The Gd5Si2Ge2 model is based on detailed first order reversal curve data, taking both reversible and irreversible properties into account, and is able to reproduce a series of independent experimental results. The Preisach models are applied to simulate material behavior under realistic application conditions in AMR-type cycles. The findings support those of the direct MCE measurements, namely that under AMR-type conditions the available MCE is bound by low field heating and high field cooling entropy curves. The heat production due to hysteresis in an AMR-type cycle initiated at a given temperature is found to be equal to that of the corresponding isothermal magnetization hysteresis loop. Furthermore the MCE is seen to be correlated to the hysteresis. This allows for relatively simple implementation of magnetic hysteresis losses in numerical AMR system models: either from measured isothermal magnetization curves or simply by adding heat production in the form of a suitably scaled MCE. Due to the history dependence of hysteretic materials, experimental procedures need careful analysis. It is demonstrated that magnetization measurements can suffer from unintended effects due uncareful change of the magnetic field and temperature. Too high magnetic field ramps in isothermal magnetization experiments can induce extrinsic hysteresis effects due to the MCE destroying the isothermal conditions, even at relatively low ramp rates. Aggressive temperature control can lead to oscillations around temperature set points, which is demonstrated to induce partial hysteresis loop behavior that will generally underestimate thermal hysteresis. Furthermore it is shown that care should be taken in non-isofield type experiments, as is the case for direct MCE experiments. Measuring the temperature dependence of the MCE can yield different results in heating or cooling modes. Cooling mode measurements will tend to be overestimated, whereas the heating mode results are representative of the realistic MCE available in application conditions.

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