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Validation of free-volume concept by using temperature-pressure equivalency of polyamide-6

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In our society, usage of polymers is increasing exponentially by replacing metals. One of the important characteristic of polymers is higher strength to weight ratio compared to metallic structures. Although, specific strength of polymers is higher than metals, but in contrast they show time-dependent mechanical properties, i.e., modulus, stiffness, elasticity and toughness. Depending on the usage conditions, stress-strain state, humidity and temperature, mechanical properties can change considerably with time. The temperature-pressure history, to which polymeric part went to while manufacturing, regulates time-dependency. A polymer comprised of free space among its molecule, and sum of all this space is termed as free volume. According to Free Volume Theory, the level of free volume will define the mechanical and thermodynamic properties of polymers, and it can be changed either by temperature or / and pressure. To predict the mechanical properties of polymers, they are normally modelled by free volume theory, i.e. Williams, Landel and Ferry (WLF) and Filler-Moonan-Tschoegl (FMT) model. Effect of temperature on mechanical properties of polymers is modelled by well known WLF equation, while combined effect of temperature and pressure is modelled by FMT equation. In order to predict polymer behaviour precisely at elevated temperature and pressure, FMT equation and consequently validity of free volume theory requires a precise experimental verification. Thermodynamically it is possible to change the volume of polymers either by temperature or by pressure, i.e. volume can be decreased either by decreasing temperature or by increasing pressure. If temperature and pressure changes simultaneously in such a way that specific volume stays same as at reference conditions. These combinations of temperature and pressure are termed as T-P equivalency in bulk behaviour. In cooling cycle of production, volume reduces, which causes shrinkage to product. By knowing T-P equivalency, one can reduce shrinkage considerably by cooling at equivalent higher pressure. Additionally, T-P equivalency can be used to analyze and optimize processing parameters critically for the sake of isotropic production in least time. As PA-6 is one of the most important polymer in engineering polymers, so some data on time-temperature and time-pressure superposition of PA-6 was available. The experimental data of this t-T-P superposition was extracted and manipulated to investigate T-P equivalency and validation of free volume concept. The analysis from experimental data disproves the free volume concept for PA-6, but the extracted experimental data was devised only for t-T-P superposition, so it has some limitations, which render this analysis unreliable. In order to confirm the validation of free volume theory, a set of new experiment were performed. In first part of this thesis, T-P equivalency for PA-6 was determined for an important range of temperature. Dilatometer insert of CEM Measuring System was used for investigation of T-P equivalency in bulk behaviour. For validation of free volume theory, an experiment was performed to measure shear relaxation modulus at equivalent temperature and pressure. According to free volume theory, mechanical properties are solely defined by content of free volume, so if volume stays constant, even at elevated temperature and pressure, the relaxation modulus should be unaltered. Shear relaxation modulus was experimentally determined by using relaxometer insert. The results from literature and experiment describe that T-P equivalency for specific volume in measured range of temperature and pressure shows a linear trend. The slope of this linear relation describes the ratio of isobaric thermal expansion coefficient and isothermal compressibility. In addition to equivalent temperature-pressure for constant specific volume, such combination of temperature and pressure were also determined that always represent constant fractional free volume. The shear relaxation modulus measured at constant fractional free volume describes that Free Volume Theory is not valid for PA-6.

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