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Temperature dependence of heterogeneous nucleation: Extension of the Fletcher model

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Recently there have been several cases reported where the critical saturation ratio for onset of heterogeneous nucleation increases with nucleation temperature (positive slope dependence). This behavior contrasts with the behavior observed in homogeneous nucleation, where a decreasing critical saturation ratio with increasing nucleation temperature (negative slope dependence) seems universal. For this reason the positive slope dependence is referred to as anomalous. Negative slope dependence is found in heterogeneous nucleation as well, but because so few temperature-dependent measurements have been reported, it is not presently clear which slope condition (positive or negative) will become more frequent. Especially interesting is the case of water vapor condensation on silver nanoparticles [Kupc et al., AS&T 47: i–iv, 2013] where the critical saturation ratio for heterogeneous nucleation onset passes through a maximum, at about 278K, with higher (lower) temperatures showing the usual (anomalous) temperature dependence. In the present study we develop an extension of Fletcher's classical, capillarity-based, model of heterogeneous nucleation that explicitly resolves the roles of surface energy and surface entropy in determining temperature dependence. Application of the second nucleation theorem, which relates temperature dependence of nucleation rate to cluster energy, yields both necessary and sufficient conditions for anomalous temperature behavior in the extended Fletcher model. In particular it is found that an increasing contact angle with temperature is a necessary, but not sufficient, condition for anomalous temperature dependence to occur. Methods for inferring microscopic contact angle and its temperature dependence from heterogeneous nucleation probability measurements are discussed in light of the new theory.