NUCLEATION MODELLING FOR SYNTHESIS OF THE NEW ADVANCED MATERIALS

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The new material design assumes to make the mixture of some components, which should be transferred into the metastable state and activated to start the chemical reactions or/and multiple phase transformations within the sophisticated technological procedures. Practically all chemical and physical treatments are presenting the energetic barriers overcoming, where new phase embryos are generating within the complicate sequence of the single steps of the new material design. Theory of nucleation is used generally to get a theoretical description and optimizing of these steps.

The first nucleation experiment is associated with measurements of liquid and crystals super cooling done by Fahrenheit (Ostwald, 1896). The nucleation of bubbles in gas saturated solutions was observed and the concept of critical embryos of a new phase was introduced in nucleation science during the second half of 19th century (Volmer, 1939). The quality of vapor/liquid nucleation rate results has improved substantially beginning in 1970s because of the development of new measurement systems. For example, the first prototype of the Flow Diffusion Chamber for vapor nucleation rate measurements was developed by Anisimov et al. (1978, 1982). Currently, the most significant problem in nucleation is the correspondence between experimental data and theoretical predictions of nucleation rate values. As a rule, theoretical and experimental data on nucleation rate are not in good agreement over a range of temperatures and/or pressures. It appears that there may be problems in both the experiments & theory and deficiencies can be identified in all versions of nucleation theories and practically all of the reported experimental results.

Current theories correspond to various modifications of Classical Nucleation Theory that was completed in the 1940s (Frenkel, 1975). The theoretical results look quite reasonable for sufficiently low vapor nucleation rates where the droplet approximation is applicable (Anisimov, 2003). However, these approaches have problems at the nanometer scale when the critical embryos contain of the order of 200 or less molecules (atoms). It appears that this quantity of molecules is near the threshold for the droplet critical embryo approximation, at least for organic vapors. Some researchers (Baydakov, 1995; Protsenko et al., 2006) have expressed optimism that nucleation theory and experiment are in agreement for the case of bubble generation from the superheated liquids.

At the present time, vapor-gas nucleation theory can produce values that deviate from the experimental results by up to several orders of magnitude (Fladerer and Strey, 2006; Brus et al., 2005). However, nucleation experiments using different devises also show significant inconsistencies in the measured rates (for example, see Brus et al., 2005). Both problems produce difficulties in establishing one or more standard vapor/liquid nucleation systems that could be used to test vapor-gas nucleation rate measurement systems. The problem of the nucleation rate standard is more complex than simply using the n-pentanol-helium system as was suggested by the International Workshop on Nucleation in the Czech Republic, Prague in 1995 as a candidate for a nucleation standard. The n-pentanol-helium system has unfortunately not produced sufficiently consistent data to date. The advantages and current problems of the vapor-gas nucleation experiments are discussed below and a view of the future studies is presented based on the assessment of vapor-gas/liquid nucleation experimental resutls.

In this review, results and problems related to aerosol generation experiments and nucleation theory development are discussed. Adiabatic expansion and gas-jet techniques, diffusion chambers, turbulent mixing apparatus were considered. It can be concluded that the development of accurate experimental techniques for vapor-gas nucleation research are still in progress. Measurable nucleation rates for the available experimental techniques span up to 19 orders of magnitude ranging from 10^{-2} $cm^{-3}s^{-1}$ up to 10^{17} $cm^{-3}s^{-1}$ and nucleation temperatures from cryogenic around 30 K to near 1300 K. Pressures in vapor-gas systems have been achieved within the interval from 30 kPa to 10 MPa. The detection of the critical vapor supersaturation for a given nucleation rate tends to be replaced by nucleation rate determinations that can vary by several orders of magnitude.

Important semiempirical information is obtained when the critical embryo parameters are derived from experimental nucleation rate data. These relationships were called “nucleation theorems.” Kashchiev (2006) recently provided a theoretical review devoted to nucleation theorems. More easily used equations to interpret experimental results have been given in several articles (Anisimov and Cherevko, 1985; Anisimov et al., 1987; Anisimov and Taylakov, 1989). Because of the inconsistencies in the experimental data, these relationship and the results derived from them should be considered as preliminary. It can be concluded now that the differences between theory and experimental data are too large to permit reconciliation of the theoretical predictions and experimental nucleation rates. Nevertheless some of the results and ideas reviewed above provide a reasonable basis for further development of the experimental vapor-gas nucleation research techniques.

It can be hoped that the uncontrolled parameter(s) will be identified in the near future and permit consistent nucleation rate data to be derived from different research methods. The introduction of one or several nucleation standard(s) is a major current problem. Success in the nucleation standard development and its introduction in nucleation research practice is a key issue for current nucleation experiments. The deeper understanding of the carrier-gas effects may clarify the nature of the different experimental set data inconsistencies. High pressure measurement systems need to be designed to permit the study of multi-channel nucleation at near critical and spinodal conditions where the nucleation rate surface topology needs to be clarified. While developing new experimental systems, the existing techniques can be used to study nucleation to explore the effects of the carrier gas and multiphase phenomena that need to be further explored. It is reasonable to attract the attention of researchers and practitioners to the new idea of semiempirical design of the nucleation rate surfaces (Anisimov, 2017). That idea can (and will) be used for data base on the nucleation rate surfaces design.

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