COATING OF AMMONIUM SALT PARTICLES BY FLUIDIZED BED PROCESSES

Alexander Dresel, Konstantin Busch, Thomas Heintz

Fraunhofer Institute for Chemical Technology, Joseph-von-Fraunhofer-Str. 7, 76327 Pfinztal, Germany

The application of ammonium salts as ammonium dinitramide (ADN) and phase stabilized ammonium nitrate (PSAN) in solid propellants is challenging. Ammonium based particles can show poor interactions with surrounding composite matrices, often exhibit unsuitable mechanical properties, low chemical stability or require adjustment of their energetic properties like for example burning behavior or impact and friction sensitivity. Fluidized bed coating is a process technology facilitating the modification of such materials by the particle encapsulation with thin homogenous coating layers. The fluidized bed process can thereby be operated to achieve a gentle coating avoiding fragmentation of brittle materials and to minimize particle agglomeration or aggregation during the modification. Improvement of particle properties can be realized by their encapsulation with polymeric systems which alter particle interactions, mechanical stability and decelerate the solubility behavior in respective fluids.

Embodied in this work is the investigation of the modification of ammonium dinitramide and phase stabilized ammonium nitrate by their encapsulation in two-component polymeric systems with a fluidized bed process. The work focused on the process development and optimization in order to achieve thin homogenous coating layers avoiding particle agglomeration and fragmentation. Thereby, a significant improvement of the fracture strength could be shown. Particularly, the influence of the particle modification on the fracture behavior could be characterized with a specific fracture strength analysis system and compared with the mechanical properties of the initial materials.

Keywords

Coating, ammonium nitrate, ammonium dinitramide, fluidized bed

1 Introduction

Ammonium dinitramide (ADN) and phase stabilized ammonium nitrate (PSAN) are propellant components for applications in defence and space. Successful application of these materials often require their modification and adjustment of their properties and material behavior as particle interaction, chemical stability, burning behavior and sensitivity. Particularly, ADN is used as ADN prills which usually exhibit low mechanical particle properties and thus, require the increase of the mechanical particle strength.

Microencapsulation of the particles can facilitate the modification of these materials [1], whereby material behavior can be improved by the combination with the properties of the coating material which is homogenously deposited on der particle surface. Challenging is thereby possible particle fragmentation of brittle materials and agglomeration during the coating process [1].

Embodied in this work is the investigation of the coating of ammonium dinitramide prills and phase stabilized ammonium nitrate with a polymeric system and the effect on the mechanical strength of the particles.

2 Materials and methods

Ammonium dinitramide prills (ADN prills, Swedish Defence Research Agency FOI, Sweden) and nickel phase stabilized ammonium nitrate (PSAN, Fraunhofer ICT, Germany) were used as core materials for the coating with polymer-systems. In particular, a two-component polyurethane coating was investigated.

The coating was achieved with a fluidized bed process. The coating system (Mycrolab, Hüttlin Germany) which operates batchwise with a particle bulk volume between 0.1 and 0.3 dm³. The coating fluid which contains the dissolved coating material is sprayed into the fluidized bed by a certain nozzle system. Thereby, the fluid wets the particles whereby the solvent evaporates driven by the heated fluidized gas flow and the coating material is deposited at the particle surface.

The particle size was analyzed with laser diffraction measurements (Mastersizer® 2000, Malvern Instruments Ltd., UK). The particles were transferred into n-heptane and dispersed by ultrasound. Three single measurements per sample were performed wherefrom an average particle size distribution was calculated. The particle morphology was characterized by a macroscope (Leica Makroskop Z16 APO with digital camera DFC420, Leica Microsystems).

The mechanical particle properties were characterized with a certain particle strength testing system (GFP-Automat, etewe, Germany). Therein, the particles are individually loaded with a

force by a tip until the particle break. The breaking strength of the particle can then be determined from the recorded force-displacement curves.

3 Results and discussion

3.1 Particle size distribution and morphology

The particle size distributions of the materials are shown in figure 1. The sizes of the initial particles (dashed lines) refer between 70 to 200 μ m and 20 to 500 μ m for the PSAN and ADN-prills respectively. In comparison to the initial particles, the coated particles exhibit bigger sizes and hence, the polymer coated PSAN and ADN-prills are in similar size region between approximately 70 μ m and maximal 1000 μ m.

The size increase is caused by the slight particle agglomeration in the fluidized bed during the coating. Thereby, particles attach to each other and do not detach before drying of the coating and remain together. The further coating process can cause the coverage of the agglomerate with coating material, the polymer, and the possible repeat of the agglomeration process by the attachment of new individual particles on the formed agglomerates. This means that the agglomeration increases with increasing process time.



Figure 1: Particle size distribution of PSAN and ADN prills before and after the polymer coating

Thereby, fine particles tendentially enhance the agglomeration which can be seen from the figure 1. The initial size distribution of the ADN-prills show a significant higher fraction of fine particles with sizes smaller than 100 μ m if it is compared with the size distribution of the

phase stabilized ammonium nitrate. The fine fraction agglomerates and cannot be detected after the coating process.

The morphology of the particles was characterized by macroscopy analyses. Macroscopy images of the particles before (a, c) and after the polymer coating (b, d) are shown in figure 2. The images show the spherical morphology of the PSAN and ADN prills whereby slight agglomeration can be observed even in the initial materials. For the initial ADN prills stronger agglomeration can be seen than in the PSAN material.

The image of the polymer coated PSAN shows the agglomerates which generally consist of several particles. In comparison to that in the ADN-prill agglomerates, small prills are preferably attached on the surface of bigger particles of sizes > 100 μ m. This means, that in the ADN material the agglomeration is dominated by the very fine particles below 100 μ m. Thus, these observations from the microscopy analyses are in good agreement with the detected particle size distributions.



Figure 2: Macroscopy images of PSAN and ADN prills before and after the polymer coating: a) PSAN, b) PSAN with polymer coating, c) ADN prills and d) ADN prills after polymer coating

However, the images of both materials – the PSAN and ADN-prills – show individual and not agglomerated particles which remain after the coating process. Furthermore, the particle surfaces of remaining individual particles as well as the agglomerate structures exhibit a homogenous coverage with the polymer. Besides that, no fragmentation during the coating process could be observed from the macroscopy images.

3.2 Effect of the coating on particle strength

The fracture strengths of the particles were determined with the strength testing system and are summarized in table 1. Particularly, in the table are the average fracture strength with standard deviation and the number of particles which were analyzed. Thereby, the given fracture strength is referred to individual particles as well as agglomerates. If it is further compared with the standard deviation it can be seen that the fracture strengths of the tested particles vary in a wide range. The strong variations are most probably caused by the following effects:

- Particle orientation and crystal structure can influence the one directed fracture force, for example ADN prills usually exhibit a layered structure [2]
- The fracture strength depends on the particle size
- Agglomerates and agglomerate structures influence the mechanical particle properties and can cause strong variations
- The layer thickness of the polymer coating varies between different particles

However, the fracture strength in table 1 indicate the tendential influence between the polymer coated and initial materials. For the PSAN no significant change can be observed in the fracture strength after the polymer coating which might be caused by the high mechanical strength of the initial PSAN as well as the strong agglomeration in that system. The formation of big agglomerates amplifies thereby the fluctuation of fracture strength. In comparison to PSAN, an increase of the average fracture strength from 11.40 to 15.98 MPa of ADN prills by the coating can be seen from the table despite the slight agglomeration and layered particle structures. This improvement of the mechanical particle properties by the ADN modification can enhance processing and application of this material.

 Table 1:
 Average fracture strength and number of test particles for PSAN and ADN prills

 before and after the polymer coating

Particles	Average Fracture Strength [MPa]	Standard deviation [MPa]	Number of tested particles [-]
PSAN	20.48	7.59	627
Polymer coated PSAN	20.60	9.35	642
ADN prills	11.40	6.20	821
Polymer coated ADN prills	15.98	9.82	937

4 Conclusion

In this work the coating of phase stabilized ammonium nitrate and ammonium dinitramide with a polymeric system could be shown. The coating could be achieved with a fluidized bed process. Thereby, a homogenous coating layer around the particle could be formed whereby slight agglomeration occurred. However, it could be shown that the polymer modification of ADN prills can lead to a significant improve of the fracture strength if it is compared with the non-coated ADN prills.

5 References

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