The Effect of Catalysts Addition to Silicone Rubber as Pressure Transmitting Medium in the Pebble Fuel Production Process

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Abstract. Pebble Fuel is a spherical fuel for high temperature gas-cooled reactors (HTGR). This fuel must have a homogeneous density distribution. Hyperelastic material is used as a pressure transmitting medium (PTM) material in making Pebble fuel using the cold quasi-isostatic pressing method. PTM material properties and characteristics were predicted using the finite element analysis method. The problem is the type of material used and its suitable composition to make a pressure-transmitting medium that has the properties and characteristics of the material as predicted. This research discusses the manufacture of tensile specimens for pressure-emitting media using RTV-586 silicone rubber. The composition comprises three different variants with two major ingredients, namely RTV-586 silicone rubber and catalyst. The test results are then analyzed using the finite element method to determine the material composition that is appropriate or close to the predicted properties and characteristics of the PTM material. This initial study used the Mooney-Rivlin hyperelastic model. The Mooney-Rivlin model shows good similarity to the test result data. In future studies, it will make comparisons with other hyperelastic models to get a suitable PTM material constant.

1. Introduction

The design of HTGR fuel elements is usually divided into two categories: prismatic fuel elements or spherical fuel elements [1]. The fabrication process of spherical fuel element is mainly composed of four steps, i.e., preparation of resinated matrix graphite powders, overcoating the Tristructural Isotropic (TRISO) coated fuel particle, cold quasi-isostatic pressing or molding, and lathing and subsequent heat treatments [2]. Cold quasi-isostatic pressing (QIP) is a pressing technique developed from the hydrostatic pressing technique. The QIP technique is efficient and simple but has the advantage of being able to provide a triaxial volumetric suppression effect on pressed material (powder), to produce a final product with a uniform density and good physical and mechanical properties [3]. The QIP method in addition to providing volumetric (triaxial) compression of the pressed material also combines the advantages of biaxial hydrostatic suppression and uniaxial static suppression [4, 5].

At present, the QIP technique in powder has gained wide acceptance in the production of ceramic, graphite, metal and abrasive components [6] as well as the fabrication process of spherical fuel elements.
for high-temperature gas-cooled reactors (HTGR). The fabrication process consists of two stages: pre-molding and final molding, where both stages use the QIP technique to reduce the anisotropic properties [7,8].

Quasi-isostatic pressing is a pressing technology for materials in powder form that allows uniform density to be obtained in all parts [9]. This process is carried out using hydraulic equipment or standard compression mechanisms. The equipment design, installation, and operational conditions for QIP are not much different from conventional compression techniques. The principle of quasi-isostatic pressing is based on the use of hyperelastic material as a medium (pressure transmitting medium) to transmit uniform static pressure in all directions. The quasi-isostatic pressing process is shown in Figure 1.

![Figure 1. Quasi-isostatic pressing (QIP).](image)

As shown in Figure 1, the process begins with the punch moving downward pressing the pressure transmitting medium (PTM) which will then transfer the pressure received to the powder to be processed. The final result that is processed depends on the nature of the PTM. If the PTM can transfer the pressure received uniformly in all directions, then the pressed powder will have a uniform density.

The PTM selection is the most important part in the compacting process with the QIP technique. PTM functions as a medium that continues isostatic pressure uniformly in all directions during the compacting process. PTM material is an elastomer which simultaneously forms part of the mold that forms the result of the compression process, providing internal and external pressure to the material being pressed [10]. Therefore, this research focuses on determining the material properties of PTM by using Finite Element Analysis (FEA) and hyperelastic models especially Mooney-Rivlin. The results obtained will be a reference in determining the material properties of PTM that will be made.

2. Material and Methods

In this section, analytic methods are presented to determine the parameters of hyperelastic materials and two types of experimental tests for hyperelastic materials, namely the uniaxial tensile test and the uniaxial compression test. For each type of test, three specimens were taken from the same RTV586 silicone rubber product with a difference in additional catalyst percentage. This is intended to investigate the composition that best suits the material characteristics needed as a PTM for the quasi-isostatic pressing process.

RTV586 silicone rubber consists of two parts, part A and part B. Where part B is the catalyst. For each catalyst composition three specimens were made for tensile and compression tests, so that there were nine test specimens. This is intended to investigate the uniformity of product compositions and to ensure that products provide representative test data.

The variation of the catalyst addition is based on the conventional RTV catalyst composition which is about 30 – 40 ml/kg (addsil RTV586 technical information). The catalyst composition of each specimen is shown in Table 1, while Figure 2 shows the entire RTV586 test specimen for tensile and compression test.
Table 1. Catalyst composition

| Specimen | RTV586 composition |
|----------|-------------------|
|          | Part A (%) | Part B (%) |
| H1-1     | 99         | 1          |
| H1-2     | 98         | 2          |
| H1-3     | 97         | 3          |
| H2-1     | 98         | 2          |
| H2-2     | 97         | 3          |
| H2-3     | 97         | 3          |
| H3-1     | 97         | 3          |
| H3-2     | 97         | 3          |
| H3-3     | 97         | 3          |

Figure 2. Tensile and compression test specimen.

The test data is used for the curve fitting process to get the RTV586 material constant and to choose the appropriate constitutive model. The requirement for the curve fitting process is a simple stress-strain function that is able to represent every expected load condition in the experiment.

In experimental testing, a Gotech AI-7000S universal servo control system and laser extensometer is used.

2.1. Uniaxial Tensile Test

In the tensile test the specimen is tested for its tensile strength by being given a tensile load to find out the maximum stress before the specimen is broken. The standard used is ASTM D412 (see fig. 3), using a universal testing machine (UTM AI-7000S Gotech) as shown in fig.4.

Figure 3. Dimensions of the tensile test specimen according to ASTM D412 [11].
2.2. Uniaxial Compression Test

In this test the specimen is tested by giving a load that presses the specimen to determine the stress and strain values that occur during loading as well as the maximum load that the specimen is able to accept. The standard used is ASTM D575-91.

Compression testing is usually done because the rubber is often applied to pressure or force when used. Compression testing is done by applying uniaxial pressure to the rubber to determine the rubber's elasticity ability when pressed.

The shape of the test specimen in the form of a cylinder refers to the ASTM standard D575-91 with the dimensions of the test specimen that is diameter 28.6 ± 0.1 mm and thickness 12.5 ± 0.5 mm (see Fig. 5).

![Figure 5. Dimensions of the compression test specimen.](image)

2.3. Hyperelastic Material Model

To study the behavior of an elastic material with a non-linear, which experienced a large strain with little emphasis given, used non-linear version of the classical elasticity theory which has been developed by Ogden [12]. The theory of non-linear elasticity, is a theoretical basis for the study of hyperelastic materials, such as silicon rubber using the strain energy function (W) to describe in the form of energy the mechanical behavior of the material.

To capture the non-linear mechanical properties of silicone rubber material in this study, a Mooney-Rivlin model will be used with three parameters of the material. It can be assumed that silicone rubber behaves like an isotropic, hyper-elastic and non-linear material [13]. The strain energy function (W) can then be described as a function of the following invariant strain:

\[
W = W(I_1, I_2, I_3)
\]

where:

\[
I_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2
\]

\[
I_2 = \lambda_1^2 \lambda_2^2 + \lambda_2^2 \lambda_3^2 + \lambda_1^2 \lambda_3^2
\]

\[
I_3 = \lambda_1^2 \lambda_2^2 \lambda_3^2
\]
where $\lambda_1$, $\lambda_2$ and $\lambda_3$ are stretching principles. Assuming that the material is incompressible ($I_3 = 1$), Equation (1) becomes:

$$W = W(I_1, I_2)$$

(3)

The Cauchy stress principle can be stated as a stretching function as follows:

$$\sigma_i = \lambda_i \frac{\partial W}{\partial \lambda_i} + p \quad i = 1, 2, 3$$

(4)

where $p$ is the hydrostatic pressure which can be determined from the boundary conditions [9]. Martins et al. [9] conducted a comparative study of several material models for the prediction of hyper-elastic properties, with applications for silicone rubber and soft tissue, using data obtained from a uniaxial tensile test.

Mooney-Rivlin was one of the first hyper-elastic models developed in 1940. Currently it is still widely used, because it has high accuracy when predicting non-linear behavior of isotropic, rubber-like material [14].

Two parameters of the Mooney-Rivlin material model in the form of a strain energy equation can be seen in the following equation:

$$W = C_{10}(I_1 - 3) + C_{01}(I_2 - 3)$$

(5)

where $C_{10}$ and $C_{01}$ are empirically determined material constants, and $I_1$ and $I_2$ are the first and second invariant unimodular components of the Cauchy-Green deformation tensor, respectively.

Whereas, for the three parameters of the Mooney-Rivlin material model in terms of strain energy can be described as follows:

$$W = C_{10}(I_1 - 3) + C_{01}(I_2 - 3) + C_{20}(I_1 - 3)^2$$

(6)

where $C_{10}$, $C_{01}$ and $C_{20}$ are an empirically determined material constant.

3. RESULTS AND DISCUSSIONS

3.1. Uniaxial Tension

Figure 6 shows the stress versus strain data for specimens with catalyst composition of 1%, 2% and 3% tested in stress. Nine silicon rubber specimens with three different catalyst compositions exhibit various behaviors under stress, highlighting their different mechanical properties (Table 3.1). The H1 specimen (H1-1 through H1-3) had the largest strain on failure (439.9%) of the three specimens. Then decrease sequentially on H2 and H3 specimens with increasing catalyst addition to the specimen.

![Figure 6. Tensile stress versus strain data as measured for 9 specimen silicone rubber RTV586 with 3 catalyst composition.](image-url)
Table 2. Tensile stress vs. strain experimental data

| No | Sample | Max Stress (Mpa) | Max Strain (%) |
|----|--------|------------------|----------------|
| 1  | H1-1   | 3,3273           | 458,3738       |
| 2  | H1-2   | 2,9089           | 421,1922       |
| 3  | H1-3   | 3,0436           | 440,1828       |
| 4  | H2-1   | 2,4271           | 309,6972       |
| 5  | H2-2   | 2,5508           | 365,0698       |
| 6  | H2-3   | 3,0463           | 373,3158       |
| 7  | H3-1   | 2,2527           | 294,8545       |
| 8  | H3-2   | 3,0999           | 327,6383       |
| 9  | H3-3   | 2,0601           | 293,4552       |

3.2. Uniaxial Compression

The stress-strain behavior of the silicon rubber sample under compression is shown in Figure 7. Displacement that occurs during the test is very small. The basic assumption of rubber behavior during this test is similar to that of fluid regarding the same reaction force in all directions. Very small volume changes during testing under hydrostatic pressure (p) characterize rubber material as a material that is virtually incompressible.

![Figure 7: Compressive stress versus strain data as measured for 9 specimen silicone rubber RTV586 with 3 catalyst composition.](image)

The deformation of nine silicon rubber samples with three different catalyst compositions, and the mechanical properties calculated under compression indicate that the compressive strain of the whole sample did not show a significant difference as shown in Table 3.
### Table 3. Compressive stress vs. strain experimental data

| No | Sample | Max Stress (MPa) | Ultimate Elongation (%) |
|----|--------|-----------------|-------------------------|
| 1  | H1-1   | 2,7092          | 65,9359                 |
| 2  | H1-2   | 2,7089          | 67,2692                 |
| 3  | H1-3   | 2,7099          | 70,8322                 |
| 4  | H2-1   | 2,7099          | 65,9055                 |
| 5  | H2-2   | 2,7098          | 63,9522                 |
| 6  | H2-3   | 2,7097          | 65,4105                 |
| 7  | H3-1   | 2,7097          | 63,0613                 |
| 8  | H3-2   | 2,7090          | 65,4162                 |
| 9  | H3-3   | 2,7090          | 67,4757                 |

### 3.3. Curve Fitting

To choose the most suitable hyperelastic model, ANSYS provides a curve fitting feature. This feature simplifies the user’s effort by comparing some available models with experimental data so we can decide on the most suitable model. Experimental stress-strain data is need to determine the exact strain energy function coefficient during the curve-fitting process. Test data needed for the installation process of the curve is a uniaxial tensile test, planar shear, equibiaxial, and volumetric (compression test) which produces an equation for the strain energy function in terms of strain invariant or hyperlastic material strain. In this research paper, the data available are only the uniaxial tensile and uniaxial compression (volumetric) test data. Therefore, curve fitting will perform by using the two data test.

The hyperelastic model used is Mooney-Rivlin with three parameters. The experimental stress-strain data was evaluation using Equation 5 to match the experimental data as shown in Figures 8 to 10. The most stable experimental data of all cycles for each test were selected for the curve fitting process [15].

![Figure 8. Curve fitting of Mooney-Rivlin model with uniaxial test data H1-1 sample.](image)
From the results of the curve fitting of the hyperelastic Mooney-Rivlin 3 model parameters, we get the PTM material constants using silicone rubber as shown in Table 3.3.

| Coef. | H1-1  | H1-2  | H1-3  | Average |
|-------|-------|-------|-------|---------|
| C_{10} | 0.2518 | 0.2394 | 0.2287 | 0.2400 |
| C_{01} | -0.2417 | -0.2321 | -0.2076 | -0.2271 |
| C_{11} | 0.0076  | 0.0077  | 0.0077  | 0.0077  |

After determining the properties of the PTM material, the next step is to simulate the quasi-isostatic pressing (QIP) process using the material properties of the hyperelastic modeling results. The QIP simulation will be carried out in future studies.
4. Conclusion and Future Works

4.1. Conclusion

The addition of a catalyst affects the mechanical properties of silicone rubber. From the experimental results, we found that the H1 sample with the addition of 1% catalyst had the best tensile and compressive stress vs strain compared to other compositions. However, the actual test data required for the curve fitting process are the uniaxial tensile test, planar shear, biaxial, and volumetric (compression test). Whereas in this study we used only the tensile test and compression test, however at least two experimental data that have been conducted can provide an initial picture in the selection of PTM materials.

4.2. Future Works

Some of the test and experimental data needed to get the exact strain energy function coefficient have not been carried out. This is because the test equipment is not available in our laboratory, so it is necessary to collaborate with other institutions. In the future, a joint testing will be carried out to complete the data so that it is possible to compare several hyperelastic models and obtain the desired PTM material coefficient.

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