3D ceramic printing of nuclear fuel

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Title **3D ceramic printing of nuclear fuel**

Author(s) / Coauthor(s): Manuel A. Pouchon* / Marco Streit**

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In this document a novel nuclear fuel production method is suggested. It combines previously developed approaches and improvements of the internal gelation process, which was used over decades to produce sphere-pac fuel for minor actinide transmutation. And modern 3D printing of ceramics. The improvements to the internal gelation, which were developed in two CCEM.CH projects in the recent years comprise the two following aspects

- Change of the heating media from silicon oil to microwave fields
- Application of in-situ preparation of the feed solution, with the sparing of any need in cooling

The internal gelation together with these improvements can be used to implement a new 3D printing technique for ceramics, and especially nuclear fuels, with the following advantages:

- Almost powder-less process up to the formation of the fuel body in complex shapes
- Any local composition change in metal or enrichment can be achieved
- Pore-formers can be introduced locally, allowing the formation of buffer regions/layers

*) also lecturer at the ETHZ and “Maître d'enseignement et de recherche” at EPFL

**) also lecturer at the ETHZ

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Author:	Signature:	Date:	Reviewer:	Signature:	Date:
Manuel A. Pouchon	[signature]	26.8.2016	Johannes Bertsch	[signature]	26.08.16

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Contents

1 Introduction

PSI has a long tradition in developing advanced nuclear fuels. Some major efforts went into the development of actinide and minor actinide containing fuels for transmutation. One route has been the development of inert matrix fuels, for plutonium transmutation in light water reactors, either as yttria stabilized zirconia pellets [1], or as CerMet pellets [2]. The other route was the development of alternative, aqueous production methods, in order to simplify the fabrication, making it ready for remote manipulation. The major effort went into the internal gelation, resulting in sphere-pac fuel. Both, the gelation and fuel performance of this particular concept are summarized in [3] and [4]. As the pellet fuel is a well experienced concept in commercial light water reactors, and also in many advanced systems, such as sodium cooled fast reactors, some efforts went also in the direction of developing aqueous fabrication routes, resulting in pellets. Early approaches concentrated on crushing spheres into pellet-shape and heat treat them (see [5] and [6]), resulting in so called hybrid pellets. But also the use of an aqueous direct ceramic shaping methods was already introduced at PSI [7], using the direct coagulation casting technique. Another effort went in the direction of freeze drying [8].

Improvements of the internal gelation process combined with an implementation of a new 3D printing technique for ceramics will enable to fabricate various complex fuels.

1.1 Motivation to New Method

This document treats a new nuclear fuel production method, providing an almost dustless fabrication method with the ability to introduce locally varying features like metal composition, enrichment and porosity. The motivations to these aspects are described in the next section.

1.1.1 Aspect of Nuclear Fuel Cycle and Nuclear Fuel Refabricating

The closure of the nuclear fuel cycle offers very promising aspects like an improved uranium resource usage and a major reduction of long lived minor actinides in the final waste. For a comparison of different aspects between the classical once through PWR scenario, the once reuse of Pu option and the fully closed fuel cycle involving fast reactors see Fig. 3 in [9].

Closing the cycle of course involves reprocessing of spent fuel, and the fabrication of highly active, minor actinide containing fuel. The high activity imposes new production challenges compared to the production of fresh nuclear fuel. Most important:

- the production must be performed remotely in a shielded cell (hot-cell), and
- any accumulation of fuel educts should be avoided.

The first aspects calls for production equipment with less maintenance need. The latter aspect disfavors powder based production, as dust is volatile and can deposit anywhere in the production environment, the hot-cell.

The solution to these challenges can be a simplified pellet process or particle fuel with much simplified production passes. At PSI the particle fuel option was researched over decades using the aqueous internal gelation process resulting in fuel pebbles used in the sphere-pac concept. An extensive description of sphere-pac and also the vipac fuel can be found in [3].

Here a new method is suggested, which makes use of the internal gelation process with some improvements, and combined with 3D printing. The aspect of simplification and reduction of dust production are hereby fully maintained.

1.1.2 Functional Fuel

Pellet fuel has heavily been optimized for UO_2 in Zircaloy cladding and LWR reactors. If considering fast reactors, other fuel matrices, such as carbides and nitrides might become more attractive because of higher metal content and better thermal conductivity. However, the swelling behavior especially of carbides is much higher compared to oxides. Therefore porosity should be designed to accommodate the dimensional change with burnup. With 3D printing and pore formers, this might be achieved locally; allowing maximal flexibility in fuel design. As Pu and minor actinides are to be introduced in fast reactor fuel for transmutation, the same applies for the local metal composition, which could be optimized with this technique.

The ATF (accident tolerant fuel) initiative is a large, post-Fukushima effort, to reduce the risk of fuel/cladding failure in case of an accident. Some of the concepts are based on oxidation and high temperature resistant ceramics. These are by nature brittle, even though pseudo ductility is introduced by using a composite. However, fuel-cladding mechanical interaction should be avoided by designing a large gap. In order to avoid an important temperature step, one concept being suggested is the introduction of porous graphite buffer. A similar feature could directly be applied by the 3D printing technique.

1.2 Base Process Already Implemented at PSI

The internal gelation technique [10] is well experienced at PSI. As illustrated above, it has been used for decades to produce pebbles for the sphere-pac concept. The main feature of the internal gelation is the heat triggered solidification process. This means that the gel can be formed by heating the feed solution, which was classically performed by hot silicon oil surrounding the droplets of feed solution. In the Swiss CCEM.CH project PINE and MeAWaT plus the European programs ASGARD [11] and PELGRIMM [12], the internal gelation was researched by using microwaves for heating [13] and later optimized producing the feed solution by in-situ mixing, in order to avoid any radiolysis and decay heating influence onto the process and to be able to work with non-cooled solutions [14]. The combination of both approaches allows a significant simplification of the production process for sphere-pac process. This approach is however also applicable for the method being suggested here.

2 Ceramic 3D Printing for Nuclear Fuel

The introduction of and improvement of 3D ceramic printing in the nuclear fuel shaping process could be done in several ways.

2.1 Existing Processes for Non-Nuclear Materials

Known ceramic printing techniques imply extrusion and powder bed technique. In the first case a paste containing the ceramic material is directly deposited to build up a 3D body. In the latter case, powder layers are deposited sequentially and partially bound by a binder or by heat treatment, such as local laser sintering. The remaining, non-bound material is removed at the end of the process. Both techniques are not useful for the production of nuclear fuel. For the extrusion technique the paste production might be a problem, as one might face blockage or inhomogeneity in composition. The latter technique is based on powder, which is not desirable in this application (as explained above), especially because a large quantity of powder has to be removed as last step in the production of the green body. A major improvement in this approach is shown in [15], where injected powder is permanently bound by laser fixing. As powder is still involved, this technique is still not ideal. For metals a new, gel based technique is suggested in [16]. For ceramics a new method is suggested using ink-jet printing of colloidal solutions [17].

In this document a similar method, but applied for fuel ceramics, is suggested. Either direct gelation using the well-known internal gelation technique, or a colloidal solution, as suggested in [17] are identified as the most promising techniques.

2.2 Internal Gelation for 3D-Printing Nuclear Fuel

The internal gelation technique (see section 1.2) has some advantageous features, which might be directly applicable to 3D printing of non-homogeneous functional fuel pellets. These are:

- With the internal gelation the feed solution would keep the low viscosity up to the point where the deposited droplet would get into contact with the heated sample and/or be exposed to a microwave field. The field could be locally applied using a coaxial antenna (similar to the coaxial microwave probe). This approach would mitigate the risk of blockage in the tubing and nozzle, which seems essential when working with nuclear fuel.

- With the in-situ mixing, the composition of the fuel could be varied with the pellet radius or with any position in the pellet. This would allow a local composition optimization of MOX pellets and/or also minor actinide containing pellets.

- The in-situ mixing would also allow the introduction of pore formers, which could be important to design the swelling behavior of advanced fuels, such as carbides, with higher swelling rates. It would potentially also allow designing foam like buffer layers at the surface, and therefore the option to minimize the fuel-cladding gap, improving the overall heat transfer and therefore lowering the fuel temperature. This could be further developed into a fuel composite avoiding extensive fuel-cladding mechanical interaction.

This production route with some additions (e.g. production of metal parts in reducing atmosphere, etc.) and application examples (pellets with composition and porosity variations, pellets with internal sealing layers, etc.) has been submitted to the European Patent Office on 23.8.2016, see [18].

2.3 Suspension based technique and other options

As suggested in [17] also a suspension based printing technique could be a good option to produce nuclear fuel. The variation on composition and porosity might be slightly more difficult, as it would imply the mixture of suspensions. Also the risk of blockage might be higher, when taking into account possible agglomerations. The drying of the suspension could similarly to the gelation be enhanced by microwave or even laser heating, similar to the method being suggested in [15].

The gelation options implying an external chemical agent, like the external gelation or the water extraction process, are probably more difficult to be implemented, as two streams should be applied and coordinated on the sample. The excess fluid, which should be removed from the sample, is probably also rather extensive, further complicating the process.

In a further developed freeze-drying process (see [19] and [8]), small droplets could be added to the frozen sample, also allowing the printing of local features.

2.4 Thermal Treatment

The calcination and sintering process is classically following the formation of the green body. However, when implying heavy densification, the geometrical shape might change due to diffusion processes, normally leading to a slight conic shape of the pellet. This would then still imply a final grinding step, which is dust intensive and requires maintenance prone equipment. This problem might be overcome by densifying each applied axial layer with laser heating. Such pre-sintered pellets could then be sintered into the final form using microwave technique, with more homogeneous lower temperatures, again mitigating deformation.

3 Conclusions

A new gelation and 3D ceramic printing based production technique for nuclear fuel is suggested in this paper. It promises a simple and dust-less process with the ability introducing local features, leading to functional fuel compacts. Aspects of mass production should be regarded later, but could represent a drawback of this approach. Parallel production using arrays could be a solution to this aspect.

Bibliography

[1] G. Ledergerber, C. Degueldre, P. Heimgartner, M. A. Pouchon, and U. Kasemeyer, “Inert matrix fuel for the utilisation of plutonium,” *Prog. Nucl. Energy*, vol. 38, no. 3–4, pp. 301–308, 2001.

[2] K. Idemitsu, “Incineration of Excess Plutonium in a CerMet Fuel,” PSI, Paul Scherrer Institut, Villigen, Switzerland, TM-43-97-29, Sep. 1997.

[3] M. A. Pouchon, G. Ledergerber, F. Ingold, and K. Bakker, “3.11 - Sphere-Pac and VIPAC Fuel A2 - Konings, Rudy J.M.,” in *Comprehensive Nuclear Materials*, Oxford: Elsevier, 2012, pp. 275–312.

[4] M. A. Pouchon, L.-Å. Nordström, and C. Hellwig, “3.25 - Modeling of Sphere-Pac Fuel A2 - Konings, Rudy J.M.,” in *Comprehensive Nuclear Materials*, Oxford: Elsevier, 2012, pp. 789–817.

[5] G. Ledergerber, “Kugeln als Ausgangsmaterial für Pellets,” PSI, Paul Scherrer Institut, Villigen, Switzerland, TM-43-89-21, Aug. 1989.

[6] G. Ledergerber, F. Ingold, R. W. Stratton, H.-P. Alder, C. Prunier, D. Warin, and M. Bauer, “Preparation of Transuranium Fuel and Target Materials for the Transmutation of Actinides by Gel Coconversion,” *Nucl. Technol.*, vol. 114, no. 2, pp. 194–204, May 1996.

[7] M. Streit, F. Ingold, L. J. Gauckler, and J.-P. Ottaviani, “(Pu,Zr)N Annular Fuel Peiets Shaped By. Direct Coagulation Casting,” *J. Nucl. Sci. Technol.*, vol. 39, no. sup3, pp. 741–744, Nov. 2002.

[8] E. Hart, “Ceramic pellet fabrication by freeze-drying,” PSI, Paul Scherrer Institut, Villigen, Switzerland, TM-43-02-02, Sep. 2002.

[9] V. Kuznetsov, G. Fesenko, A. Schwenk-Ferrero, A. Andrianov, and I. Kuptsov, “Innovative Nuclear Energy Systems: State-of-the Art Survey on Evaluation and Aggregation Judgment Measures Applied to Performance Comparison,” *Energies*, vol. 8, no. 5, pp. 3679–3719, Apr. 2015.

[10] M. A. Pouchon, “14 - Gelation and other innovative conversion processes for aqueous-based reprocessing and recycling of spent nuclear fuels A2 - Taylor, Robin,” in *Reprocessing and Recycling of Spent Nuclear Fuel*, Oxford: Woodhead Publishing, 2015, pp. 353–369.

[11] “FP7 EU project: Advanced fuelS for Generation IV reActors: Reprocessing and Dissolution (ASGARD) (Project reference: 295825).” [Online]. Available: http://cordis.europa.eu/project/rcn/100635_en.html. [Accessed: 18-Aug-2016].

[12] “FP7 EU project: PELlets versus GRanulates: Irradiation, Manufacturing & Modelling (PELGRIMM) (Project reference: 295664).” [Online]. Available: http://cordis.europa.eu/project/rcn/101413_en.html. [Accessed: 18-Aug-2016].

[13] M. A. Pouchon, “PINE - Platform for Innovative Nuclear Fuels,” in *CCEM :: Annual Activity Report 2012*, Paul Scherrer Institut, Villigen, Switzerland: CCEM.CH, pp. 39–40.

[14] M. A. Pouchon, “MeAWat - Methods of Advanced Waste Treatment,” in *CCEM :: Annual Activity Report 2014*, Paul Scherrer Institut, Villigen, Switzerland: CCEM.CH, pp. 52–54.

[15] Z. Liu, K. Song, B. Gao, T. Tian, H. Yang, X. Lin, and W. Huang, “Microstructure and Mechanical Properties of Al2O3/ZrO2 Directionally Solidified Eutectic Ceramic Prepared by Laser 3D Printing,” *J. Mater. Sci. Technol.*, vol. 32, no. 4, pp. 320–325, Apr. 2016.

[16] X. Ren, H. Shao, T. Lin, and H. Zheng, “3D gel-printing—An additive manufacturing method for producing complex shape parts,” *Mater. Des.*, vol. 101, pp. 80–87, Jul. 2016.

[17] M. Bienia, M. Lejeune, M. Chambon, V. Baco-Carles, C. Dossou-Yovo, R. Noguera, and F. Rossignol, “Inkjet printing of ceramic colloidal suspensions: Filament growth and breakup,” *Chem. Eng. Sci.*, vol. 149, pp. 1–13, Jul. 2016.

[18] M. A. Pouchon, “Aqueous additive production method for the fabrication of ceramic and/or metallic bodies,” Application Number at European Patent Office: EP16185359.3, Filing Date: 23 August 2016.

[19] M. Morihira, “Application of Freeze-drying Process for Spherical Fuel Fabrication,” PSI, Paul Scherrer Institut, Villigen, Switzerland, TM-43-99-36, Aug. 1999.

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