

Imaging Nylon Polymerisation Processes by Applying Electrical Tomography

T. Dyakowski¹, Michal Mikos², Dimiter Vlaev¹, Reg Mann¹, G.W.Follows³
Arthur Boxman³ and M.P.W. Wilson⁴

¹Department of Chemical Engineering, UMIST, PO Box 88, Manchester M60 1QD United Kingdom

²Institute of Heat Engineering, Warsaw University of Technology

³DuPont (U.K.) PO Box 401, Middlesbrough, TS6 8JE

⁴DuPont (U.K.) Ltd, Gloucester GL4 3HP

¹The authors are members of the Virtual Centre of Industrial Process Tomography

Abstract – *The paper presents an application of electrical resistivity tomography for imaging a nylon polymerisation process at elevated temperatures (up to 275 °C) and pressures (up to 22 bar). The process was realised within a metal walled tank using a sensor constructed in the form of a ceramic sleeve with electrodes coated on the inner surface. The images obtained show the dynamic behaviour of the process and how the electrical properties of the material varied with time.*

Keywords: electrical tomography, nylon polymerisation, and plasma technology

1. INTRODUCTION

Electrical tomography has been applied to imaging electrically conducting media (resistivity tomography) or dielectric media (capacitance tomography) at ambient temperature and pressure [1]. These applications were mainly concerned with imaging flow morphology of multi-phase flows: gas–solids (capacitance tomography) and solids–liquid (resistivity tomography). Non-homogeneities in phase distribution resulting from various types of interactions between phases were imaged and those images provided valuable information on the dynamic behaviour of multi-phase flow [2]. The results were obtained for flow conditions characterised by constant physical parameters (conductivity or permittivity). For such conditions the measured signals were *only* functions of material distribution in a pipe or a vessel cross-section. For example, for resistivity tomography, this significantly simplified the measurement protocol by using only one set of injecting currents.

This paper presents an application of electrical tomography, which is focused on imaging a nylon 66 polymerisation process. A key difference from earlier work is that the electrical properties of the material studied varies significantly during this process, the first step of which involves reacting hexamethylene diamine and adipic acid at low temperature to form hexamethylene diammonium adipate or nylon 66 “salt”. In the laboratory, polycondensation of this salt to polymer is carried out with a 85% salt : 15% water solution in a stainless steel autoclave

and involves pressures approaching 20 bar and temperatures up to 300°C.

The polymerisation process itself maybe divided into four stages which can be described as heating, boiling, pressure reduction and equilibrium. During the first two stages, condensation commences but because of the high pressure the reaction mixture remains quite fluid and low molecular weight products remain in solution, albeit they become more and more viscous as the condensation proceeds. During third stage, pressure reduction, heat continues to be applied to raise the temperature close to 275°C whilst simultaneously reducing pressure. After reaching atmospheric pressure the batch temperature is maintained at ca.275°C to complete the polymerisation after which the molten polymer is extruded from the bottom of the autoclave.

It will be appreciated that as the condensation proceeds there is a progressive removal of water with a consequential significant impact on electrical properties.

2. EXPERIMENTAL FACILITIES

A key challenge at the outset of this work was to obtain tomographical images within a metal-walled pressure vessel. This has been achieved with the set-up illustrated in Figures 1a - 1c. To insulate the electrodes from the autoclave metal wall, a ceramic sleeve with a set of 16 electrodes was inserted into the autoclave, Figures 1a and 1b. Spraying a composite powder (80% nickel and 20% chromium) onto the sleeve inner

surface formed the electrodes. This was achieved by applying a plasma technique at atmospheric pressure, the composite powder being introduced into the plasma jet, the inlet temperature of which was 15,000 °C and the outlet temperature 8,000°C. Solid particles were melted and liquid particles hit the inner surface with the velocity of about 300 ms⁻¹; to assure good adherence, the inner surface of the ceramic sleeve was rough. Additionally, a cooling system was applied to control the rate of solidification of melted particles.

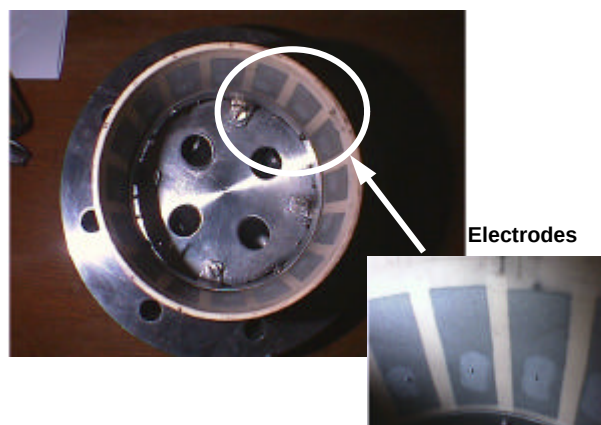


Figure 1b: Ceramic sleeve (φ120) with 16 embedded electrodes

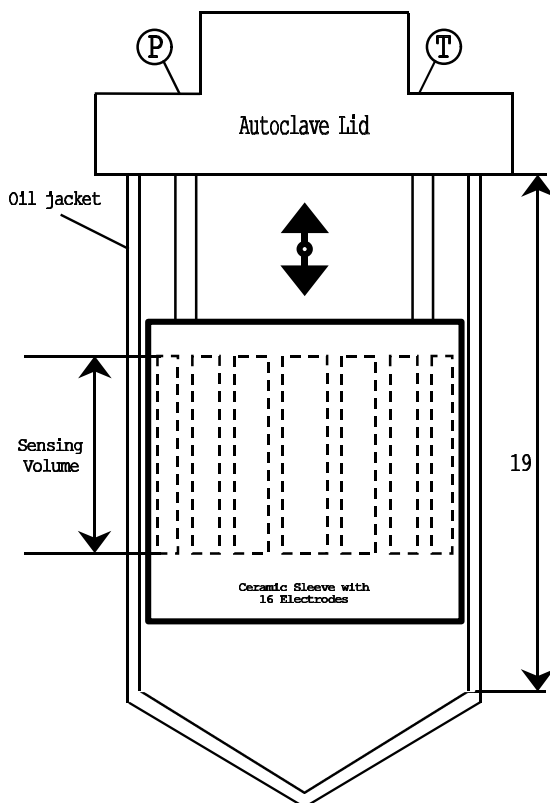
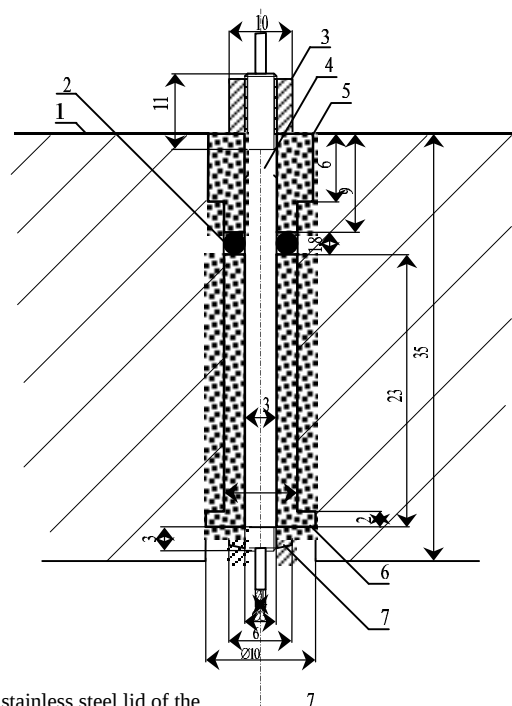


Figure 1a: Ceramic sleeve design allowing suspension at various vertical positions

The sensor was attached to the autoclave lid in which 16 extra holes were made for the wires conducting the electrical signal from the 16 electrodes to the data acquisition system. Both pressure and electrical insulation were required for those holes, so special non-conductive and heat resistant materials were used (Figure 1c). The shoulder washers (5 and 6) were manufactured from PEEK and were used to support the metal elements while providing good electrical insulation. The o-rings were used to seal the 17 bar pressure and were made from Kalrez®(2), a special DuPont insulating material whose mechanical properties do not undergo change in the temperature range experienced during nylon polymerisation.



- 1 - stainless steel lid of the autoclave
- 2 - Kalrez® o-rings
- 3 - stainless steel nut
- 4 - stainless steel rod
- 5,6 - peek shouldered washer
- 7 - stainless steel nut

Figure 1c: Pressure and heat proof embedded tomography sensors

A 0.5 mm K-type thermocouple applicable in the range -50 to 1300 °C was inserted in a 3-mm stainless steel tube which was fitted in an additional 17th hole in the lid.

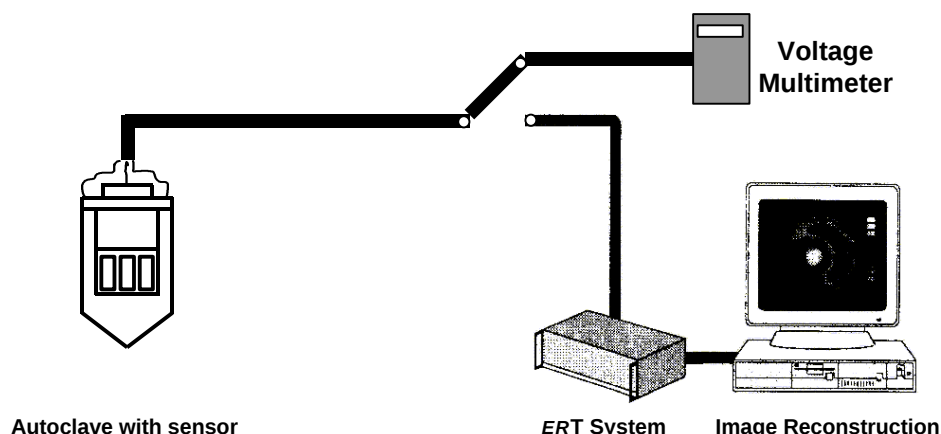


Figure 1d: Experimental set-up providing tomographic and direct measurement of solution conductance

At the start of the heating stage, the mixture of salt and water is non-conductive but subsequently conductivity increases as the nylon salt dissolves in the water. However at the end of the boiling process when nearly all of the water of solution has been removed, conductivity again approaches a value close to zero. Such variations in conductivity imposed some requirements concerned with the measurement protocol. Thus, in addition to the ERT system an independent electronic circuit was applied to measure only local variations in conductivity between adjacent and opposite pairs of electrodes, Figure 1d. The conductivity measurements obtained from this circuit were applied for the initial set up of the ERT system as well as for changing it during the imaging process. This covered both the changes of the injection current (from 1mA to 15 mA) as well as adjusting the gain map ('U' shape curve).

3. RESULTS AND DISCUSSION

The results displayed were obtained during two consecutive tests conducted at the DuPont laboratory at Wilton. First, simply imaging boiling water and its transformation into steam tested the sensor assembly. These tests were conducted up to 210 °C at pressures up to 17 bar. Changes in the conductivity scale were observed when pressure was sharply reduced at high temperature, which caused vigorous boiling. No pressure leakage or damage to the equipment was observed.

In practice, the dry nylon 66 salt is fed into the autoclave together with a small amount of water (85% nylon and 15% water). It takes time for the water to penetrate through the entire volume of the nylon salt and to dissolve it, which is facilitated by increasing temperature during the heating up stage. As the salt is dissolved, the region where this occurs becomes conductive. This is illustrated by the conductance variation between the adjacent and opposite pairs of

electrodes (Figure 2). For the adjacent pair of electrodes, this occurs after about 30 minutes from the start of the heating up stage, taking a little longer for the total volume as indicated by the results from the opposite pair of electrodes. The variation in average conductance, calculated from 104 voltage measurements obtained from a set of 16 electrodes, is shown in Figure 3. The tomographic data obtained during the second run showed a very good degree of repeatability as Figure 3 indicates. As also illustrated in Figures 2 and 3, the average conductivity started to decline a little before the maximum pressure of 17 bar was reached.

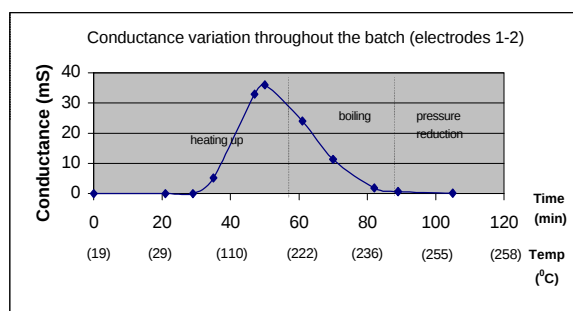


Figure 2a: Conductance variation throughout the batch measured between 2 adjacent electrodes

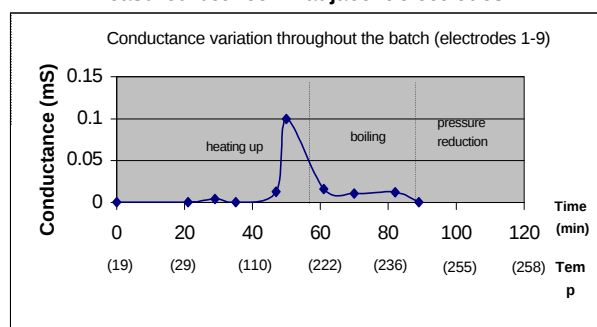


Figure 2b: Conductance variation throughout the batch measured between 2 opposite electrodes

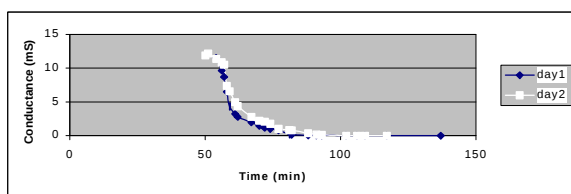


Figure 3: Conductance variation with time during two days of nylon polymerisation

Within the heating up stage, the ERT system was set up with an injection current of 15 mA. The images presented in Fig 4 show little variation, which corresponds to a nearly uniform distribution of the material within the vessel. During the following stages the injection current was adjusted according to the changes in the conductance as shown in Table 1.

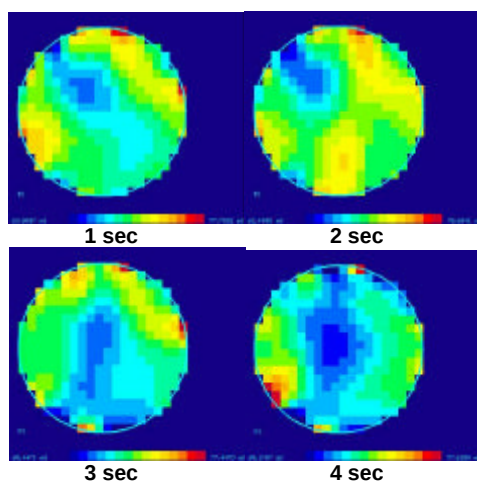


Figure 4: The first images obtained in a nylon producing autoclave (heating up)

Time (min)	45	72	86	97	110
Current (mA)	15	10	5	3	1

Table 1: Injected current variations

It was observed that the boiling and pressure reduction stages were accompanied by more non-homogeneous material distribution than during the heating up stage, Figure 5. During the boiling stage additional heat is applied and the pressure control valve bleeds off steam to maintain constant pressure. This results in drastic changes of resistance distribution as illustrated in Figure 6.

4. CONCLUSIONS

The paper presents the first steps in the application of resistivity tomography for monitoring a chemical process at elevated temperatures and pressures. A specially designed sensor assembly enabled electrical signals to be taken from a metal walled vessel and through a metal lid. Plasma technology was

utilised to fabricate a set of electrodes on the inner surface of the ceramic sleeve inserted into the metal walled autoclave and tests showed that the electrodes could withstand operating conditions within the autoclave.

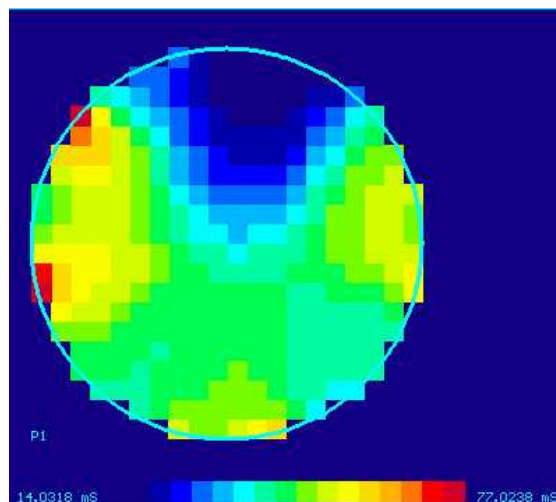


Figure 5: Stable zone of low conductivity observed during nylon polymerisation (boiling)

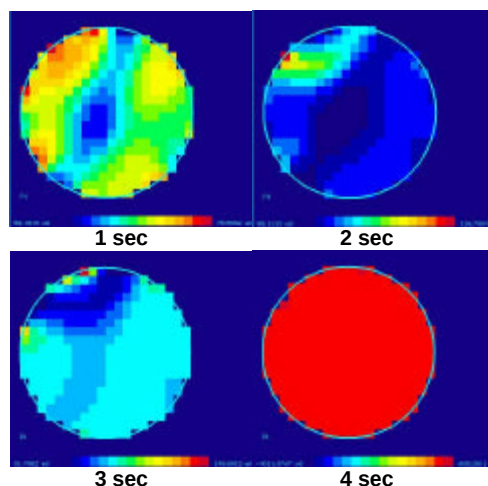


Figure 6: Drastic changes in the images coinciding with the valve bleeding off (pressure reduction)

The results obtained illustrate how the electrical conductance varies during the various stages of the nylon 66 polymerisation process. Imaging started at the end of the heating up stage and continued through the boiling and pressure reduction stages showing how material is distributed. The effect of opening the pressure control valve during the boiling stage was detectable.

The technique provides the basis for obtaining useful information about important stages of a nylon polymerisation process. In addition, future application of either impedance measurements or solely capacitance tomography would permit imaging at the beginning of the heating stage and during final equilibrium.

ACKNOWLEDGEMENTS

The authors wish to express their thanks to Mr Michael Slinger from the laboratory at DuPont Wilton for his assistance during experimental trials.

The authors acknowledge the support of EPSRC and DTI through grant GR/L 37434.

REFERENCES

- [1] R.A. Williams and M.S. Beck, Eds *Process Tomography: Principles, Techniques and Applications*, Butterworth Heinemann, Oxford, 1995.
- [2] T. Dyakowski, "Process Tomography for Multi-phase Measurement", *Measurement Science and Technology*, **7**, pp. 343-353, 1996.